



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7 :

C09J 7/02, 5/06, B60J 10/02, B29C
65/34, B32B 27/08

A1

(11) International Publication Number:

WO 00/27941

(43) International Publication Date:

18 May 2000 (18.05.00)

(21) International Application Number:

PCT/US99/24255

(22) International Filing Date:

15 October 1999 (15.10.99)

(30) Priority Data:

09/189,045

9 November 1998 (09.11.98)

US

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Box 33427, Saint Paul, MN 55133-3427 (US).(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG,
BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE,
ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP,
KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA,
MD, MG, MK, MN, MW, MX, NC, NZ, PL, PT, RO, RU,
SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG,
UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS,
MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ,
BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE,
CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC,
NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA,
GN, GW, ML, MR, NE, SN, TD, TG).

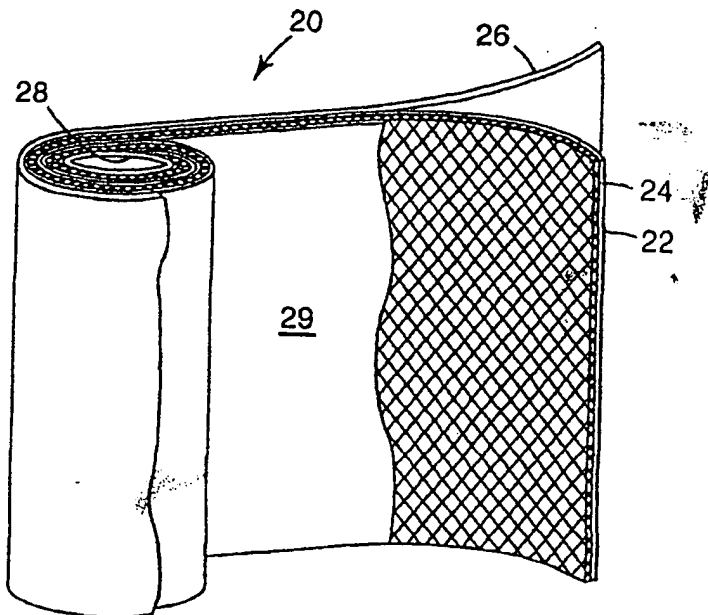
Published

With international search report.

(54) Title: BENDABLE ARTICLE CONTAINING A HEATING ELEMENT, ASSEMBLIES MADE THEREWITH, AND A METHOD
OF USING SAID ARTICLE

(57) Abstract

The invention relates to articles, including tapes that comprise a heating element and a polymeric material. In a preferred embodiment the polymeric material is heat-activatable, to establish a seal or a bond between two substrates, particularly where at least one of the substrates is glass.



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5 BENDABLE ARTICLE CONTAINING A HEATING ELEMENT, ASSEMBLIES
MADE THEREWITH, AND A METHOD OF USING SAID ARTICLE

Field of the Invention

10 The invention relates to articles, including tapes that comprise a heating element and a polymeric material. In a preferred embodiment the polymeric material is heat-activatable, to establish a seal or a bond between two substrates, particularly where at least one of the substrates is glass.

Background of the Invention

15 Many applications exist where it is necessary to "join" together two substrates using adhesives and/or sealants to create a bond and/or a seal. A "bond" refers to uniting two substrates, which may be the same or different, with an adhesive material between them. A "seal" refers to uniting two substrates with a sealant to exclude air and/or water from passing between the substrates. A sealant can also function as an adhesive.

20 Some substrates are particularly difficult to adhere to. Such substrates include ceramic materials such as glass and glass frit and thermoset materials such as thermoset composite materials and cured paints. The difficulty is compounded when trying to join two different substrates such as glass and painted metal. For example, glass windshields are bonded within the metal or plastic frame of a motor vehicle both during vehicle
25 manufacture and following manufacture to replace the windshield in the event that it cracks or breaks. Additionally, a seal must be established between the windshield and the frame to prevent water from leaking into the vehicle. To enhance adhesion, the glass surface is typically primed prior to inserting it into the frame.

30 Polyurethane pastes are conventionally used to establish a seal between the primed glass and the frame. Such pastes, however, are difficult to apply uniformly and reproducibly. The pastes often contain solvents and have reactive isocyanate materials which are environmentally undesirable. Additionally, these compositions cure with moisture from the

atmosphere so that a relatively long time is required to cure and build bond strength. During this vulnerable curing period, the glass can vibrate within the frame, making the seal and the glass susceptible to damage. Gaps in the seal can form, giving rise to wind noise and compromising seal integrity. The noise associated with the vibrations is also undesirable.

5 Moreover, the reliance on ambient moisture means that the cure process varies depending upon ambient conditions.

EP 0262831 (Martin) describes a method for heat curing an adhesive using an electrical resistor such as a resistance wire. The wire is adhered to the frame in a rebate and a ribbon of a polyurethane sealant is applied from a dispensing cartridge over the wire. The ribbon is flattened when the windshield is placed over the rebate. The wire is then connected
10 to a power source to generate heat to cure the composition.

U.S. Patent No. 4,184,000 (Denman) describes a coupling device for glass or metal parts. The coupling device is used to attach or remove a windshield to an opening in an automotive vehicle. The coupling device has a body and an inner core having a heating
15 element which can be a simple wire, a wound wire, a foil tape, or an impregnated core. Upon heating, only the outer portion of the body becomes fluidly adhesive while the rest of it remains plastically deformable under the applied load. In a preferred embodiment, the heating element is a wave formed wire which improves the heat distribution.

U.S. Patent No. 4,555,607 (Roentgen et al.) describes a glass pane having a
20 conductive strip around its marginal edge that can be heated to treat a strand of adhesive for adhering a glass pane to a frame, and also for removing of the glass pane. The conductive strip can be a silver baking paste that is silk screened onto the glass and then burnt in during a heat treating process, or it can be a metal applied by flame spraying or vacuum metallizing.

25 Adhesives having electrically conductive heating elements are also described in, for example U.S. Patent Nos. 3,049,465 (Wilkins), 5,100,494 (Schmidt), 3,438,843 (Pagel), Other methods of bonding materials using electrical heating elements include U.S. Patent No. 5,389,184 (Jacaruso et al.)

30 Summary of the Invention

The present invention provides an article comprising:

a laminate, the laminate comprising:

(a) a heating element selected from the group consisting of:

(i) an electrically conductive mesh;

5 (ii) a component comprising a continuous electrically conductive path, the path having a plurality of thermally conductive appendages attached to the continuous electrically conductive path in such a manner that heat generated in the continuous path by electricity flowing therethrough can be thermally conducted through the appendages;

(b) a polymeric material;

wherein the article is capable of passing Coplanar Arc Bend Test A1.

10

In one embodiment of the article of the invention the polymeric material comprises a heat activatable material selected from the group consisting of thermosettable materials, thermoplastic materials, and mixtures thereof, wherein the heat activatable material is in thermal contact with the heating element, such that if the heat activatable material is not in
15 direct contact with the heating element it is separated from the heating element by a material that is thermally conductive to a sufficient degree to allow activation of the heat activatable material, and wherein the heating element is capable, upon having an electrical current flow therethrough, of causing at least a portion of the heat activatable material to soften, melt, and/or cure.

20

In one embodiment of the article of the invention the polymeric material comprises a conformable, compressible melt flow-resistant thermoset core.

In one preferred embodiment of the article of the invention the article is capable of
25 passing Coplanar Arc Bend Test A1, and even more preferably also Coplanar Arc Test A2.

30

In one embodiment of the article of the invention the heating element is capable of passing Coplanar Arc Bend Test A2.

In one embodiment of the article of the invention the heating element is capable of passing Coplanar Arc Bend Test A3.

In one embodiment of the article of the invention the heating element is capable of passing Coplanar Arc Bend Test B1, preferably also Coplanar Arc Test B2, and even more preferably Coplanar Arc Bend Test B3.

5 In one embodiment of the article of the invention the article is capable of forming a water tight seal.

 In one embodiment of the article of the invention the heating element is embedded in the heat activatable material.

10

 In one embodiment of the article of the invention the heating element comprises a mesh.

 In one embodiment of the article of the invention the mesh is a substantially rectangular piece of mesh having a first major surface and an opposing second major surface.

15

 In one embodiment of the article of the invention the heating element is substantially rectangular.

20

 In one embodiment of the article of the invention the heat activatable material is directly attached to a side of the heating element.

 In one embodiment of the article of the invention the heat activatable material is selected from the group consisting of sealants and adhesives.

25

 In one embodiment of the article of the invention the adhesive is a hot melt adhesive.

 In one embodiment of the article of the invention the heat activatable material is continuous.

30

In one embodiment of the article of the invention the heat activatable material is in the form of a substantially rectangular strip.

5 In one embodiment of the article of the invention the article is in the form of a substantially rectangular strip.

In one embodiment of the article of the invention the heating element is equal to or less than the width of the heat activatable material.

10 In one embodiment of the article of the invention the mesh is selected from the group consisting of knit mesh, braided mesh, pierced and expanded mesh, pierced and expandable mesh, and mesh on a bias.

15 In one embodiment of the article of the invention the heating element is the component of (a)(ii).

In one embodiment of the article of the invention the appendages of the component are in substantially the same plane.

20 In one embodiment of the article of the invention some of the appendages are attached to and extend away from one side of the path and a remainder of the appendages are attached to and extend away from an opposite side of the path.

25 In one embodiment of the article of the invention each appendage attached to the path is spaced less than about 1 cm from another appendage.

In one embodiment of the article of the invention each appendage attached to the path is spaced within about 5 mm from another appendage.

30 In one embodiment of the article of the invention the article further comprises a conformable, compressible melt flow resistant thermoset core.

In one embodiment of the article of the invention the laminate further comprises a conformable, compressible melt-flow resistant thermoset core, wherein the heating element is positioned between the heat activatable material and the core.

5 In one embodiment of the article of the invention the laminate further comprises a conformable, compressible melt flow resistant thermoset core, wherein the heat activatable material is positioned between the heating element and the core.

10 In one embodiment of the article of the invention the heating element is embedded in the heat activatable material and wherein the laminate further comprises a conformable, compressible melt flow resistant thermoset core.

In one embodiment of the article of the invention the core comprises a foam core.

15 In one embodiment of the article of the invention the core is inherently tacky.

In one embodiment of the article of the invention the laminate further comprises a layer of adhesive coated on one or both sides of the core.

20 The present invention also provides an assembly comprising:

- (a) a first substrate;
- (b) an article of the invention joined to the first substrate.

25 The assembly optionally further comprises a second substrate and wherein the first substrate and second substrate are joined together via the article.

30 In an embodiment of the assembly of the invention the first substrate is selected from the group consisting of vehicle glazing, architectural glazing, computer screens, television screens, vehicle body panels, carpeting, and flooring.

In one embodiment of the assembly of the invention the first substrate comprises a windshield.

In one embodiment of the assembly of the invention the first substrate comprises a windshield and the second substrate comprises a vehicle frame.

5 In one embodiment of the assembly of the invention the first substrate is selected from the group consisting of glass, metal, painted metal, primed metal, polyurethane, plastics (thermosets, thermoplastic, thermosetting), wood, ceramics, minerals, and masonry.

The present invention also provides a method comprising the steps of:

- 10 (a) providing a first substrate;
- (b) providing a stack of materials in contact with the first substrate, wherein the stack of materials comprises:
- (i) a heating element selected from the group consisting of:
- 15 (A) an electrically conductive mesh, wherein the mesh is capable of passing Coplanar Arc Bend Test B1;
- (B) a component comprising a continuous electrically conductive path, the path having a plurality of thermally conductive appendages attached to the continuous electrically conductive path in such a manner that heat generated in the continuous path by electricity flowing therethrough can be thermally conducted through the appendages, and
- 20 wherein the component is capable of passing Coplanar Arc Bend Test B1;
- (ii) a heat activatable material selected from the group consisting of thermosetting materials, thermoplastic materials, and mixtures thereof, wherein the heat activatable material is in thermal contact with the heating element, such that if the heat activatable material is not in direct contact with the heating element it is separated from the heating
- 25 element by a material that is thermally conductive to a sufficient degree to allow activation of at least a portion of the heat activatable material, and wherein the heating element is capable upon having an electrical current flow therethrough of causing at least a portion of the heat activatable material to soften, melt, and/or cure;
- (c) placing a second substrate against an exposed surface of the stack; wherein
- 30 alternatively the stack is simultaneously provided between the first and second substrate;
- (d) causing an electrical current to flow through the heating element during one or more of the following: step (b), step (c), after step (c); in a manner to cause the heat activatable

material to soften, melt, and/or cure such that the first substrate is ultimately joined to the second substrate through the stack in order to form an assembly.

5 In one embodiment of the method of the invention the stack of materials are in laminate form prior to step (d). (For example, the stack of materials are in the form of a tape.)

10 In one embodiment of the method of the invention the stack further comprises a conformable, compressible, melt flow resistant thermoset core; and optionally further comprises one or more of the following: an adhesive layer, a primer layer, a tie layer.

15 The method of the invention optionally further comprises the step (e) of causing an electrical current to flow through the heating element to cause some or all of the laminate to soften and/or melt in order to separate the first substrate from the second substrate, wherein optionally mechanical force may be used in addition to facilitate separation of the first substrate and second substrate.

20 In one embodiment of the method of the invention the first substrate is a window and the second substrate is a window frame or the first substrate is a window frame and the second substrate is a window.

In one embodiment of the method of the invention the first substrate is a window and the second substrate is vehicle window frame or the first substrate is a vehicle window frame and the second substrate is a window.

25 The present invention also provides an assembly comprising:
(a) a first substrate;
(b) a second substrate;
(c) a laminate joining the first and second substrate together wherein the laminate comprises a

30 (i) a heating element selected from the group consisting of:
(A) an electrically conductive mesh, wherein the mesh is capable of passing Coplanar Arc Bend Test B1;

(B) a component comprising a continuous electrically conductive path, the path having a plurality of thermally conductive appendages attached to the continuous electrically conductive path in such a manner that heat generated in the continuous path by electricity flowing therethrough can be thermally conducted through the appendages, and wherein the component is capable of passing Coplanar Arc Bend Test B1;

(ii) a polymeric material.

In one embodiment of the assembly the polymeric material is selected from the group consisting of heat activated sealant, heat activatable sealant, heat activated adhesive, and heat activatable adhesive.

In the assembly of the invention the laminate may optionally further comprises a conformable, compressible, melt flow resistant core, and wherein the heating element is a mesh and wherein the first substrate is a window and the second substrate is a window frame.

In an embodiment of the assembly of the invention the polymeric material is in thermal contact with the heating element, such that if the polymeric material is not in direct contact with the heating element it is separated from the heating element by a material that is thermally conductive to a sufficient degree to allow upon having an electrical current flow therethrough of causing the polymeric material to soften and optionally melt.

In an embodiment of the assembly of the invention the polymeric material is selected from the group consisting of thermoset polymeric material and thermoplastic material.

In an embodiment of the assembly of the invention the first substrate is a windshield and the second substrate is a vehicle window frame.

The term "laminate" as used herein refers to materials that are attached (via an adhesive or sealant, for example and/or mechanically attached via clips or staples, for example. Typically however it refers to materials that are bonded and/or sealed together.

5 Brief Description of the Drawings

Fig. 1 illustrates a cross-sectional view of an article of the invention.

Fig. 2 illustrates a perspective view of another embodiment of the invention which is in the form of a roll.

10 Fig. 3 illustrates a cross-sectional view of another embodiment of the invention.

Fig. 4 illustrates a cross-sectional view of another embodiment of the invention.

Fig. 5 illustrates a cross-sectional view of another embodiment of the invention.

Fig. 6 illustrates a cross-sectional view of another embodiment of the invention.

Fig. 7 illustrates a cross-sectional view of another embodiment of the invention.

15 Fig. 8 illustrates a cross-sectional view of another embodiment of the invention.

Fig. 9 illustrates a perspective view of another embodiment of the invention.

Fig. 9a illustrates a top plan view of the article of Fig. 9 as it is being bent.

Fig. 10 is a top plan view of a piece of conductive mesh that can be cut on a bias to provide a useful heating element for use in the article of the invention.

20 Figs. 11a-e are top plan views of different embodiments of useful heating element for use in the article of the invention wherein each heating element is a component comprising an electrically conductive path and thermally conductive appendages.

Fig. 11f is a top plan view of a narrow piece of conductive mesh that can be stretched if desired to provide a useful heating element for use in the article of the invention which is a component comprising an electrically conductive path and thermally conductive appendages.

25 Fig. 12 illustrates top plan view of a piece of conductive mesh which is useful as a heating element in an article of the invention.

Fig. 12b is a top plan view of a heating element useful according to the present invention which is a conductive knit mesh.

30 Fig. 12b is a top plan view of a heating element useful according to the present invention which is a conductive knit mesh.

Fig. 13a illustrates a front plan view of an assembly of the invention comprising a windshield having an article of the invention attached around its perimeter which is attached to a power source.

5 Fig. 13b illustrates a front plan view of another embodiment of an assembly of the invention comprising a windshield having an article of the invention attached around its perimeter which is attached to a power source.

Fig. 13c illustrates a front plan view of an another embodiment of an assembly of the invention comprising a windshield having an article of the invention attached around its perimeter which is attached to a power source.

10 Fig. 14 is a cross-sectional view taken along line 14-14 on Fig. 13a.

Fig. 15 is a schematic view which helps illustrate the Test Methods which help measure how easily an article of the invention can bend around an arc.

15 Fig. 16 is a perspective view an assembly of the invention comprising a window having a article of the invention around the perimeter thereof which is in position for placement into a vehicle.

Fig. 17 illustrates a partial perspective view of a piece of conductive mesh which would be useful as a heating element in an article of the invention.

Detailed Description of the Invention

20

Article

The invention is a typically elongate article for establishing a seal and/or bond between two substrates. In particular, the article is useful for establishing a seal and/or a bond between substrates in which the article must be bent into a coplanar arc between the two
25 substrates. For exemplification, the invention will be described as a tape having a substantially rectangular cross-section, although other forms are contemplated, including sheets, and tapes having other cross-sections such as circular, oval, trapezoidal, and the like. However, although the article of the invention is described frequently herein as a tape the features, properties, etc. of the tape of the invention are applicable to the article of the
30 invention in its broadest sense which is not necessarily in the form of a tape unless indicated otherwise. A sheet refers to the article in a flat form having a length, width, and thickness. A tape refers to a narrower strip which is longer than it is wide. In one embodiment the tape 10

(Fig. 1) features an electrically conductive heating element 12 which is a mesh that is capable of being bent into a coplanar arc as defined below and at least one layer 14 of a polymeric material. The tape 10 is also capable of being bent into a coplanar arc.

In one embodiment of the invention, the polymeric material is a heat activatable material. As used herein, the term heat activatable material includes thermoplastic materials, thermosetting materials, and blends thereof. Further, it means that upon heating, the material will soften and/or melt if it is a thermoplastic material; or it will soften and cure, melt and cure, or cure if it is a thermosetting material or a blend of thermosetting and thermoplastic materials. The term "thermoplastic" means that the material is capable of being repeatedly heated and cooled, respectively causing the material to soften and harden through a temperature range typical of the material, i.e., below its degradation temperature. The term "thermosetting" means that the material is capable of being changed into a substantially infusible or insoluble material when cured by heat or other means. The term "thermoset" means that a material has been cured by heat or other means and is substantially insoluble and infusible.

The heat activatable materials are preferably sealants and/or adhesives. A "sealant composition" or a "sealant layer" is a gap-filling material. Consequently, at the time of seal formation, sealant compositions useful according to the invention typically have a rheology such that the sealant composition is able to flow into and fill gaps in the substrate to which it is applied and, after the sealant has cured (in the case of thermosetting sealant compositions) or solidified upon cooling (in the case of thermoplastic sealant compositions), still sufficiently fill the gaps so as to seal the substrate. Sealant compositions useful in the invention are preferably non-tacky (i.e., they are not tacky to the touch at room temperature) once they have cured (in the case of thermosetting sealant compositions) or solidified upon cooling (in the case of thermoplastic sealant compositions). Preferably, the sealants useful according to the invention also function as adhesives in that they bond to the surfaces that they seal.

In a second embodiment of the invention, the polymeric material comprises a conformable, compressible, melt flow resistant core as described below. In a highly preferred embodiment, the tape comprises a heat activatable layer, an electrically conductive heating element, and a conformable, compressible, melt flow resistant core.

In the practice of the invention, the article may optionally be conveniently provided in the form a roll of tape 20 (Fig. 2) for easy storage, shipping, handling and use. In one construction, the tape 20 comprising a sealant layer 29, an electrically conductive heating element 24 which is a mesh, and an adhesive layer 22 is typically wound about an optional core 28. Core 28 can be paper or plastic, for example, and typically has an inside diameter about 7.6 centimeters. In such constructions, the tape may be wound up with a temporary, removable liner 26 that separates adjacent windings in the roll. The provision of the tape in roll form is facilitated by selecting the polymeric layer such as a sealant layer to have a thickness and a modulus that promotes easy wind-up without exerting a force that could result in permanent deformation in the article, oozing of any of the layers in the article beyond the widest layer in the article, or telescoping of the roll article, when stored under ordinary, ambient conditions of temperature and humidity.

In a second construction, the tape comprises a sealant layer, an electrically conductive heating element, and optionally, a second sealant layer on the other side of the heating element. In a third construction, the tape comprises an adhesive layer, an electrically conductive heating element, and optionally, a second adhesive layer on the other side of the heating element. In a preferred construction, the tape 30 (Fig. 3) comprises a sealant layer 31, an electrically conductive heating element 24, a conformable, compressible, melt flow resistant core layer 32, and optionally, an adhesive layer (not shown). An optional, temporary, removable liner 39 may be included on the tape, especially if the core layer 32 and/or sealant layer 31 have a tacky surface at room temperature, i.e., at about 23°C. The liner is removed prior to attaching the surface that it protects to a substrate. The tape may have more than one core layer and/or sealant layer as well as other layers which include, for example, a primer or tie layer(s), adhesive layer(s), and nonwoven scrim(s). More than one heating element may also be included in the tape. Other layers that are added should not inhibit the efficacy of the electrical heating element or prevent bending of the tape as described below.

The electrically conductive heating element may be positioned anywhere within the tape construction. For example, in a tape 40 (Fig. 4) comprising a sealant layer 31, a core layer 32, and an electrically conductive mesh heating element 24, the heating element 24 is disposed on the surface of sealant layer 31. The heating element may be partially embedded into the surface of the sealant layer (not shown), or heating element 24 may be embedded

completely into the surface of sealant layer 31 of tape 50 which also includes core layer 32 (Fig. 5). Heating element 24 (Fig 6A) may also be embedded within the sealant layer 61 of tape 60, which also includes a first core layer 66, a second core layer 68, and a liner 39.

Functionally, this embodiment could be achieved by laminating together two layers of the same or different sealant compositions with the electrically conductive heating element disposed between the two sealant layers. Instead of being embedded in sealant the heating element may optionally be positioned between two separate sealant layers. Additionally, multiple heating elements may be used; the elements may have the same or different electrical resistivity.

The tape of the invention can be used to seal or bond a variety of substrates together. The substrates may be the same or different. They may be different in size, shape, or composition, for example. Examples of suitable substrates include those made of glass, metal, plastic, wood, masonry, and ceramics. Representative plastic substrates include polyvinyl chloride, ethylene-propylene-diene monomer rubber, polyurethanes, polymethyl methacrylate, engineering thermoplastics (e.g., polyphenylene oxide, polyetheretherketone, polycarbonate), sheet molding compounds, and thermoplastic elastomers, including thermoplastic elastomeric olefins. Glass, and polymers that may be used as substitutes for glass (e.g., polycarbonate and polymethyl methacrylate), may be referred to as glazing materials. The surface of the substrate may be coated, e.g., with paint, an abrasion-resistant coating, or an anti-glare coating. In the case of, e.g., windshields, the glass may include a ceramic-frit layer.

The tape is useful for sealing glass substrates to, e.g., metal, painted or primed metal, and plastic substrates. For example, the tape is useful for sealing a glass windshield to a frame (such as a metal or plastic frame, for example) in a motor vehicle. The tape is particularly useful for sealing a replacement window to the frame of a motor vehicle. In this situation, the tape may need to bond to the residual sealant, typically a polyurethane, that may be left on the frame as well as painted or primed metal areas where the sealant may have been removed.

In the practice of the invention, the tape is capable of being bent into a coplanar arc i.e., a curved line segment. By "coplanar arc" it is meant that a tape, defined as the material between a first plane, i.e., three points, on a first major surface of the tape and a second plane on the second major surface of the tape and substantially parallel to the first

plane, can be bent laterally such that the tape will substantially remain between the two planes. For a more detailed explanation of these terms and the methods involved please see the Test Methods. The tape is deemed bendable in a coplanar arc if, after bending it can be affixed to a flat surface, e.g. with a clip or pressure-sensitive adhesive tape, such that the first plane is the flat surface and the second plane is on the surface of the tape parallel to the first plane, and the tape prior to bending would have substantially been in the same planes. For example, when a rectangular piece of tape 130 (Fig. 9) having electrically conductive heating element 102 on a polymeric material 101 is bent laterally such that the outer edge 122 of tape 130 extends further than the inner edge 121, the tape 130A (Fig. 9a) conforms to the shape two substantially concentric arcs, an outside arc 122A formed by outer edge 122, and an inside arc 121A formed by the inner edge 121 such that the outside arc is greater than the inside arc. Preferably, the tape 116 (Fig. 15) is capable of being bent onto a coplanar arc with the inside edge 106 around an arc α of at least 90 degrees around a circle 105 having a radius of 10 cm. More preferably, the tape (not shown) is capable of being bent into a coplanar arc around a 180 degree arc of a circle having a radius of 10 cm, and even more preferably, the tape is capable of being bent around a coplanar arc of 90 degrees around a circle having a radius of 4 cm. If necessary, the tape can be heated in an oven or with a hot air gun to soften the tape so that it is sufficiently malleable to conform to the arc. It is noted that if the heat activatable material is thermosetting, it is desirable to keep the temperature of the tape below the thermosetting temperature until it is heated for its end use. A small amount of buckling or wrinkling of the tape, as visible to the unaided eye, is acceptable as long as the tape performs its intended function after it has been heat activated. For example, the tape may have small wrinkles in it from bending around the arc. However, upon heat activation the tape flows out, eliminating the wrinkles and forming a gap filling watertight seal on a substrate.

If the cross section of the tape is not rectangular, e.g., triangular, the first plane is in the base of the triangle and the second plane is the plane tangent to the highest point or apex of the triangle and parallel to the first plane. For a tape having a circular cross section, the first plane is tangent to one edge of the tape and the second plane is the plane tangent to the opposite edge of the tape and parallel to the first plane.

The ability of the tape to bend into a coplanar arc is largely dependent upon the rigidity of the electrically conductive heating element as well as the ability of the heating

element to stretch or elongate at fairly low forces as will be discussed below. (i.e. the ability of the heating element to bend into a substantially coplanar arc.) The tape preferably exhibits the same low force to elongate at some temperature below the melting and/or curing temperature of the polymeric material. When the heating element is too rigid or stiff, any amount of heating will not overcome the deficiencies of this rigidity when the tape is bent into a coplanar arc. For example, a tape with a rigid heating element such as a welded screen when subjected to the substantially coplanar arc bend test can exhibit lifting of the outer edge of the tape and/or buckling and kinking on the inside edge of the tape thus causing it to fail the test. A tape with a narrow ribbon heating element can potentially exhibit twisting or deformation of the heating element. Additionally, a narrow ribbon will not distribute heat uniformly across the width of the tape resulting in cold spots and inadequate sealing or bonding as would the heating element useful according to the present invention. A flat foil (wider than a ribbon) can tear or lift on the outside edge because that edge is being forced around a radius that is longer than its inside radius. Multiple wires will tend to bunch up and pull in toward the inside edge of the arc causing non-uniform heating and possibly a short circuit. The wires may remain coplanar, or they may pop out of the plane of the tape to compensate for the greater distance that outside wires need to bend on an arc.

The electrically conductive heating element useful in the invention comprises at least one electrically conductive path and appendages that are thermally conductive and may be electrically conductive, and that can be bent to form a coplanar arc. When the electrically conductive heating element is bent into a coplanar arc on an adhesive tape, it is flexible enough so that any buckling, rippling or lifting of the edges can be pushed back onto the tape into a substantially coplanar arc. Examples of suitable heating elements include conformable meshes, i.e., structures having a network of open spaces, and conformable elongated structures having appendages extending laterally therefrom, i.e., a single elongate structure with thermally conductive appendages attached to it.

In a preferred embodiment, the electrically conductive heating element is a conductive mesh 102 (Fig. 9). The connected portions 112 of the mesh provide an electrically conductive path while optional appendages 125 extend from the conductive path and provide heat to the areas of the tape in which they are in thermal contact. In an alternative embodiment the mesh of Fig 12 the outermost extensions 127 of the mesh 123 are

considered thermally conductive appendages which are also electrically conductive. Number 128 indicated the current flow direction

Similar to the definition of a coplanar arc for a tape described above, the electrically conductive heating element is defined as the material between two parallel planes, each characterized by three points, on two parallel surfaces of the electrically conductive heating element, and a coplanar arc is formed by laterally bending the electrically conductive heating element. The electrically conductive heating element is coplanar if any appendages, ripples or kinks can be lightly pressed onto and affixed to the tape. For example, a rectangular strip of the heating element 102 in Fig. 9 in the form of a diamond-shaped mesh has lateral edges 151 and 152, which are defined by the straight edges of the tape. When the mesh is bent laterally into a coplanar arc, lateral edge 152 forms an outer edge of 152A (See Fig. 9a) of the bent mesh. The outer edge is an arc of a circle having a radius of R (not shown). Similarly, lateral edge 151 forms an inner edge 151A (Fig. 9a) of the bent mesh. The inner edge is an arc of a circle concentric with the circle of the outer edge and having a radius of $(R - W)$ wherein R is the radius of the outer arc and W is the width of the mesh. The mesh 102A (Fig. 9a) has been bent laterally into a coplanar arc without the above-described deficiencies of a heating element that is too rigid. In actuality, when the mesh is bent, lateral edge 152A expands and lateral edge 151A compresses so slight deformations that can occur are acceptable. For example, the wires of the mesh can overlap, they can become slightly kinked from handling, the unattached outside extensions 125 can get bent out from between the two planes defining the mesh. The numbered items in Fig. 9 have the same numbers as in Fig. 9a except for the addition of the letter "A" thereafter. These deformations are typically less than about 7 times the thickness of the mesh, preferably, less than about 5 times. Preferred electrically conductive heating elements are sufficiently flexible such that when they are attached to a conformable pressure-sensitive adhesive tape (such as 3M 4941 available from 3M Co.), the tape with the heating element can be bent into a coplanar arc.

The width of the mesh may also deform slightly from attempting to bend it into a substantially coplanar arc. Typically the wires of the mesh can pull together slightly causing a decrease in the width of the mesh. Acceptable narrowing of the mesh or necking down is less about than 15 percent of the original width, preferably less than 10 percent, and more preferably less than about 7 percent. For example, a mesh having a width, defined as the distance between its two lateral edges, of 1.27 cm could decrease in width by no more than

about 0.2 cm after it has been bent. Preferably, the mesh, in the form of a strip, is flexible enough to form a coplanar circle having an inside radius of 10 cm.

The meshes useful for the invention are typically sufficiently flexible that they can expand and/or compress in a longitudinal direction. When a strip of the mesh is formed into an arc such that the inside edge of the mesh is aligned with an arc drawn on a surface, the outside edge of the mesh must expand; in some cases, the inside edge may also compress. Alternatively, when the outside edge of the mesh is aligned on the circle, the inside edge must compress while the outside edge may or may not expand.

The preferred meshes exhibit a tensile and elongation curve having a yield stress indicated on a stress strain curve as having high elongation under low stress. Typically, under a force of one pound (4.45 Newtons), a 0.635 cm (1/4 inch) wide mesh should elongate more than about 0.5%, and preferably more than about 1% and more preferably more than about 3%. Under a force of 2 pounds (8.90 Newtons) the aforementioned mesh should elongate more than about 1%, preferably more than about 3%. It is believed that the useful meshes elongate initially because the wires are straightening out and the stretch can be effected by a relatively low force. Once the wires of the mesh have straightened out, the mesh stretches very little until breaking strength of the wires is reached. It is the relatively high amount of stretch at a low force allows the mesh to be bent into a coplanar arc.

The meshes preferably should not be constrained by more than one wire running substantially parallel to the lateral edges of the tape or they may fail the substantially coplanar arc bend test. When two or more of the wires of a mesh 100 Fig. 10 are substantially parallel with respect to the tape, the parallel longitudinal wires 107 of the mesh have the same effect as separate parallel wires in that they tend to bunch up if they are not welded together, and/or may cause lifting of the mesh from the tape when the tape is bent into an arc thus causing it to fail the substantially coplanar arc bend test. In Fig. 10 the cross wires are identified as 105. Additionally, the longitudinal wires are already aligned so that when pulled, the elongation is limited by the tensile strength of the wires and the outside edge of the mesh cannot elongate sufficiently under low force to be bent into a coplanar arc. Likewise, constraining wires, bus bars and the like at the lateral edges of the mesh will inhibit bending into a coplanar arc.

However, when a mesh with longitudinal wires 100 (Fig. 10) is cut or formed at an angle or on a bias so that the lateral edges 109 of the mesh are ultimately aligned with the lateral edges of the tape, the longitudinal wires 107 are no longer parallel to the lateral edges

of the tape and can expand and/or compress as needed to bend into a coplanar arc. Additionally, meshes having crimped wires parallel to the lateral edges of the tape can be suitable because the crimping allows the wires to stretch to form a coplanar arc.

5 The term "electrically conductive mesh" as used herein refers to a material having multiple continuous electrically conductive paths.

Examples of meshes useful for the electrically conductive heating element include, but are not limited to, pierced and expanded meshes such as pierced and expanded conductive metal foils or polymeric films, pierced and expandable meshes such as pierced and expandable conductive metal foils or polymeric films, woven cylindrical meshes, woven
10 screen or meshes on a bias, welded screen on a bias, nonwoven meshes such as nonwoven carbon fibers and steel wool, braided conductors such as braided cable conductors, circular braid conductors, flat braid conductors, and knit mesh. An example of a pierced and expanded conductive metal foil is shown in Fig. 12 and of a knit mesh 3 is shown in Fig. 12b. The arrow 4 in Fig. 12b and arrow 128 in Fig. 12 show the direction of current flow.

15 It is not required that the meshes be rectangular in its outer dimensions. The outside edges may be for example, irregular, scalloped, and the like. However, when the mesh is attached to a rectangular conformable pressure-sensitive adhesive tape, the mesh on the tape can be bent into a substantially coplanar arc.

In a highly preferred embodiment, the useful meshes are pierced and expanded or
20 pierced and expandable metal foils. Such foils are available from Delker Corp. and Exmet Corp. In these foils, the metal is pierced with slits that cross the width of the foil. The foil is then expanded in the lengthwise or longitudinal direction to form the openings of a mesh. The openings of the mesh are defined by a mesh size having an LWD and an SWD. An opening 170 (Fig. 17) is defined by an LWD 176 which is the long axis way of the diamond,
25 measured in inches or mm from the center of the joint to the center of the adjoining joint. The mesh size is also defined by SWD 178, which is the short way axis of the diamond, measured in inches or mm from the center of the joint to the center of the adjoining joint. The mesh opening is also characterized by a thickness 172 and a strand width 174. The SWD is determined by the amount of expansion in the foil. Typically, the SWD of a mesh is about
30 0 to about 100 percent of the LWD. Preferably, the SWD is about 25 to 75 percent of the LWD. Typically the LWD should be less than the width of the mesh or the mesh is no longer a mesh as it can lose its multiple circuits. Typically, the width of the mesh and the tape

article are about the same and the width of the tape is about 1 to 10 times the LWD, preferably about 2 to about 8 times the LWD, and most preferably about 3 to about 5 times the LWD. Typically the LWD is substantially perpendicular, preferably perpendicular to both of the two long outer edges of the tape. A low value of LWD may be advantageous with respect to conductivity and melting of the heat activatable material but it may be more difficult to bend the mesh into a coplanar arc.

An alternative embodiment of the electrically conductive heating element is a component comprising a single electrically conductive path, having appendages attached thereto. The electrically conductive path is an elongate structure such as a wire or a narrow foil or film, a serpentine shaped wire, and the like, as long it is sufficiently flexible to be bent in a substantially coplanar arc and the appendages can be held down with an adhesive tape. The appendages can be in any desired shape including, for example, rectangular, triangular, and tubular. Examples of this embodiment are shown in Figs. 11a-11f. Figs. 11a-e are top plan views of different embodiments of useful heating element for use in the article of the invention wherein each heating element is a component comprising an electrically conductive path 111 and thermally conductive appendages 118, 116, 114, 112, and 113 respectively.

Fig. 11f is a top plan view of a narrow piece of conductive mesh that can be stretched if desired to provide a useful heating element for use in the article of the invention which is a component comprising an electrically conductive path 110 and thermally conductive appendages 115.

The appendages do not need to be uniform, i.e., some may be shorter while others are longer, there may be triangular ones as well as rectangular ones on the same path. The spacing and placement of the appendages may also vary. For example, all of the appendages can lie on one side of the path, they can be placed directly opposite to each other along the path, or they may alternate from one side to the other along the path. Spacing of the appendages may depend on the end use desired. Typically, the appendages may be less than 10 mm apart, and more typically less than 5 mm apart. The appendages are capable of conducting heat from the electrically conductive path to the extremities of the appendages. The appendages may also be electrically conductive. The appendages can be formed, for example, from metal wires such as those made from copper, steel, nichrome, nickel, aluminum, and the like, or ceramic materials such as aluminum oxide.

The electrically conductive heating element can be made from any material that has a sufficient electrical resistance to generate sufficient heat to effect the sealing and/or bonding desired. The useful range of resistivities will vary depending upon the power, voltages, and current to be applied, generally taking into account safety factors. The specific resistivity for a certain application is selected based on such factors as, for example, the tape dimensions (length, width, and thickness), the heat capacity of the heat activatable material, the requisite heat activation temperature of the tape, and the ambient temperature at which the tape is heated. Suitable materials for the electrically conductive heating element include, for example, metals such as copper, nickel, steel, aluminum, etc.; metal alloys such as nichrome, stainless steel, etc.; conductive polymer compositions such as conductive polymers, polymers filled with conductive materials, and blends thereof, carbon fibers, etc. Preferred materials, such as nickel, are those that do not change in electrical resistivity due to corrosion or environmental factors, e.g., humidity and heat

Typically, the outside, i.e., lateral edges of the heating element such as a mesh are coextensive with the outside, i.e. lateral, edges of the tape. However, the heating element can be narrower than the rest of the tape construction as long as sufficient heat is generated by the electrical current to activate the heat activatable material as desired. The heating element may also extend beyond the lateral edges of the tape.

In a preferred embodiment, tape 30 (Fig 3) comprises a core layer 32, a sealant layer 31, an electrically conductive heating element 24, and an optional liner 39. One purpose of core layer 32 is to act as an integral spacer when the tape is used to establish a seal or a bond between a pair of substrates. Thus, during pressurized application of the tape-bearing substrate to the other substrate, core layer 32 prevents the two substrates from coming together in the event that the sealant is displaced. Such contact is particularly undesirable where one of the substrates is glass because the resulting stress can cause the glass to break. The core layer also distributes forces resulting from cure of the sealant, thereby minimizing stress in the seal.

The core layer also preferably acts as an internal vibration damper to minimize noise associated with variable frequency substrate movement once the two substrates have been sealed together. The core layer may also isolate one substrate to which it is affixed from forces transmitted to that substrate and from the other substrate e.g., in the case of a glass

windshield installed in a motor vehicle, the core layer damps vibrations arising from wind impinging on the glass, as well as vibrations arising from the motor vehicle frame.

5 A core layer may thermally insulate the sealant layer from an adhesive layer, regardless of whether the adhesive layer is integral with the tape, or applied separately to the substrate surface prior to application of the tape. In this way, the respective curing reactions that may take place in the sealant and adhesive layers can be isolated from each other, affording the opportunity to cure the tape in stages. It also offers the advantage of increasing formulation freedom with respect to the compositions of the sealant and adhesive layers.

10 To achieve these functions a core layer is designed to be compressible and conformable. These features enable a core layer, to cushion the substrate to which the tape is affixed, and to absorb energy and distribute forces applied to the sealed construction. In addition, compressibility and conformability aid in achieving complete body contact and seal formation.

15 Core layer 32 is also designed to be melt-flow resistant such that it does not undergo macroscopic mass flow when exposed to the temperatures and pressures used during the sealing operation.

20 The thickness of a core layer should preferably be sufficient for the core layer to perform the bond line spacing function and, preferably, the vibration damping and thermal insulation functions as well. The particular thickness of a given core layer is selected based upon the application for which the tape is intended. For example, in the case of motor vehicle windshield installation, the thickness of the core layer must be small enough such that the tape can fit within the frame for which the windshield is designed. Typically, the thickness of a core layer is at least about 1 mm, preferably at least about 2 mm, and more preferably at least about 3 mm. Alternatively, the core layer can be a single stratum, or it
25 may have multiple strata.

Preferred materials for a core layer are viscoelastic materials. These materials may be thermoplastic or thermoset, with thermoset materials being preferred. Thermoplastic materials should have a melt temperature above the heat activation temperature of the heat activatable materials in the tape. Examples of suitable materials for core layer 32 include
30 thermoset materials such as crosslinked polyacrylates and polyurethanes, and thermoplastic materials such as ethylene-vinyl acetate copolymers. Examples of preferred, commercially

available polyacrylate materials are 3M™ Acrylic Foam Tapes such as 4959, 4941, 4910 sold by 3M Company.

5 Polyurethane-based core layers can be provided as solid elastomers or as cellular foams and may be formed from one- or two-part compositions. One-part compositions can be moisture-activated, in which case water, either purposefully introduced or acquired from the atmosphere, initiates the curing reaction. Alternatively, a blocked isocyanate may be used with heat being employed to unblock the isocyanate and initiate the curing reaction. Two-part urethanes comprise a first component that comprises one or more isocyanate-based resins and second component that comprises one or more polyols and
10 curatives.

Also suitable are pressure sensitive adhesives. Such adhesives allow the free ends of the tape to be fused together in the form of a joint to yield a continuous seal, preferably a joint in which the tape ends remain in the same plane such as a side-to-side joint, scarf joint, or butt joint. In addition, when a core layer is in the form of a pressure sensitive adhesive, it is
15 possible to bond the core layer directly to the substrate, thereby eliminating the need for a separate adhesive layer (integral or otherwise).

Preferably, a core layer is in the form of a foam, with thermoset acrylic foams being particularly preferred. The foam may have an open or closed cell structure, although closed cell foams are preferred. Polyethylene and ethylene vinyl acetate-based foams may also be
20 used and are typically produced by extruding a resin composition from an extruder and foaming the material before or after crosslinking. Commercial suppliers for these types of foam include Voltek Div. of Sekisui America Corp., Lawrence, MA or Sentinel Products Corp., Hyannis, MA.

Other materials that can be incorporated into core layer 32 during manufacture
25 include, for example, stabilizers, antioxidants, plasticizers, tackifiers, flow control agents, adhesion promoters (e.g., silanes and titanates), colorants, thixotropes, and other fillers.

Additionally, one or more layers of adhesive may be laminated to the core. This is particularly useful if the core itself is not adhesive in nature. An adhesive layer can be a pressure-sensitive adhesive, a pressure-activated adhesive, a hot melt adhesive film, a
30 thermosetting adhesive, or an adhesive that is a blend of a thermosetting and thermoplastic material.

Referring to tape 30 (Figure 3), sealant layer 31 is preferably in the form of a continuous layer. However, a discontinuous sealant layer may also be used as long as the sealant fuses under the application of heat and pressure to form an effective seal in the final article. To aid in achieving a good seal to irregular surfaces, the surface of sealant layer 31 available for sealing to the second substrate may be textured. In addition, both single and multi-layer sealant compositions are envisioned. The width of sealant layer 31 is application-dependent. In general, however in the form of a tape, the width of sealant layer 31 is no greater than the width of core layer 32.

The purpose of sealant layer 31 is to establish and maintain a seal between a pair of substrates. Melt-flowable compositions may be used for sealant layer 31. Suitable compositions include thermosettable materials such as epoxy resins, or the combination of such materials with thermoplastic materials, such as for example polyacrylates and polyesters to form miscible or physical blends.

One suitable class of materials includes blends of epoxy resins with semi-crystalline polymers such as polyesters. Semi-crystalline polymers are advantageous because they contribute to rapid build-up of sealant strength, leading to a seal having good green strength.

A polymer that is "semi-crystalline" displays a crystalline melting point, as determined by differential scanning calorimetry (DSC), preferably with a maximum melting point of about 200 °C. Crystallinity in a polymer is also observed as a clouding or opacifying of a sheet that had been heated to an amorphous state as it cools. When the polymer is heated to a molten state and knife coated onto a liner to form a sheet, it is amorphous and the sheet is observed to be clear and fairly transparent to light. As the polymer in the sheet material cools, crystalline domains form and the crystallization is characterized by the clouding of the sheet to a translucent or opaque state. The degree of crystallinity may be varied in the polymers by mixing-in any compatible combination of amorphous polymers and semi-crystalline polymers having varying degrees of crystallinity. The clouding of the sheet provides a convenient non-destructive method of determining that crystallization has occurred to some degree in the polymer. During use when the preferred sealants based on blends of epoxy-containing material and polyester components softens, flows and fills gaps in the surface to be sealed, the epoxy resin and the polyester component form a homogenous system as evidenced by a lack of macroscopic phase separation to the unaided human eye.

The polymers may include nucleating agents to adjust the rate of crystallization at a given temperature, and thus the rate at which green strength builds. Useful nucleating agents include microcrystalline waxes. A suitable wax is, for example, sold by Petrolite Corp. as Unilin™ 700.

5 Preferred polyesters are hydroxyl-terminated and carboxyl-terminated polyesters that are semi-crystalline at room temperature. Other functional groups that may be present include -NH, -CONH, -NH₂, -SH, anhydride, urethane, and oxirane groups.

The preferred polyesters are also solid at room temperature. Preferred polyester materials have a number average molecular weight of about 7,500 to 200,000, more
10 preferably from about 10,000 to 50,000, and most preferably, from about 15,000 to 30,000.

Polyester components useful in the invention comprise the reaction product of dicarboxylic acids (or their diester equivalents, including anhydrides) and diols. The diacids (or diester equivalents) can be saturated aliphatic diacids containing from 4 to 12 carbon atoms (including branched, unbranched, or cyclic materials having 5 to 6 carbon atoms in a
15 ring) and/or aromatic acids containing from 8 to 15 carbon atoms. Examples of suitable aliphatic diacids are succinic, glutaric, adipic, pimelic, suberic, azelaic, sebacic, 1,12-dodecanedioic, 1,4-cyclohexanedicarboxylic, 1,3-cyclopentanedicarboxylic, 2-methylsuccinic, 2-methylpentanedioic, 3-methylhexanedioic acids, and the like. Suitable aromatic acids include terephthalic acid, isophthalic acid, phthalic acid, 4,4'-
20 benzophenonedicarboxylic acid, 4,4'-diphenylmethanedicarboxylic acid, 4,4'-diphenylthioetherdicarboxylic acid, and 4,4'-diphenylaminedicarboxylic acid. Preferably the structure between the two carboxyl groups in the diacids contain only carbon and hydrogen, and more preferably, the structure is a phenylene group. Blends of the foregoing diacids may be used.

25 The diols include branched, unbranched, and cyclic aliphatic diols having from 2 to 12 carbon atoms. Examples of suitable diols include ethylene glycol, 1,3-propylene glycol, 1,2-propylene glycol, 1,4-butanediol, 1,3-butanediol, 1,5-pentanediol, 2-methyl-2,4-pentanediol, 1,6-hexanediol, cyclobutane-1,3-di(2'-ethanol), cyclohexane-1,4-dimethanol, 1,10-decanediol, 1,12-dodecanediol, and neopentyl glycol. Long chain diols including
30 poly(oxyalkylene)glycols in which the alkylene group contains from 2 to 9 carbon atoms, preferably 2 to 4 carbon atoms, may also be used. Blends of the foregoing diols may be used.

Useful commercially available hydroxyl terminated polyester materials include various saturated linear, semi-crystalline copolyesters available from Hüls America, Inc. such as Dynapol™ S330, Dynapol™ S1401, Dynapol™ S1402, Dynapol™ S1358, Dynapol™ S1359, Dynapol™ S1227, and Dynapol™ S1229. Useful saturated, linear amorphous copolyesters available from Hüls America, Inc. include Dynapol™ S1313 and Dynapol™ S1430.

Useful epoxy-containing materials are epoxy resins that have at least one oxirane ring polymerizable by a ring opening reaction. Such materials, broadly called epoxides, include both monomeric and polymeric epoxides and can be aliphatic, cycloaliphatic or aromatic.

These materials generally have, on the average, at least two epoxy groups per molecule (preferably more than two epoxy groups per molecule). The "average" number of epoxy groups per molecule is defined as the number of epoxy groups in the epoxy-containing material divided by the total number of epoxy molecules present. The polymeric epoxides include linear polymers having terminal epoxy groups (e.g., a diglycidyl ether of a polyoxyalkylene glycol), polymers having skeletal oxirane units (e.g., polybutadiene polyepoxide), and polymers having pendent epoxy groups (e.g., a glycidyl methacrylate polymer or copolymer). The molecular weight of the epoxy-containing material may vary from 58 to about 100,000 or more. Mixtures of various epoxy-containing materials can also be used.

Useful epoxy-containing materials include those which contain cyclohexene oxide groups such as the epoxycyclohexanecarboxylates, typified by 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, 3,4-epoxy-2-methylcyclohexylmethyl-3,4-epoxy-2-methylcyclohexanecarboxylate, and bis(3,4-epoxy-6-methylcyclohexylmethyl) adipate. For a more detailed list of useful epoxides of this nature, reference may be made to U.S. Patent No. 3,117,099.

Further epoxy-containing materials which are particularly useful are glycidyl ether monomers such as glycidyl ethers of polyhydric phenols obtained by reacting a polyhydric phenol with, e.g., an epichlorohydrin (e.g., the diglycidyl ether of 2,2-bis-(2,3-epoxypropoxyphenol)propane). Further examples of epoxides of this type which can be used in the practice of this invention are described in U.S. Patent No. 3,018,262. Other useful glycidyl ether based epoxy-containing materials are described in U.S. Patent No. 5,407,978.

There are a number of commercially available epoxy-containing materials which can be used. In particular, epoxides which are readily available include octadecylene oxide, epichlorohydrin, styrene oxide, vinylcyclohexene oxide, glycidol, glycidyl methacrylate, diglycidyl ether of Bisphenol A (e.g., those available under the trade designations EPON SU-8, EPON SU-2.5, EPON 828, EPON 1004F, and EPON 1001F from Shell Chemical Co., and DER-332 and DER-334, from Dow Chemical Co.), diglycidyl ether of Bisphenol F (e.g., ARALDITE GY281 from Ciba-Geigy), vinylcyclohexene dioxide (e.g., ERL 4206 from Union Carbide Corp., Danbury, CT), 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexene carboxylate (e.g., ERL-4221 from Union Carbide Corp.), 2-(3,4-epoxycyclohexyl-5,5-spiro-3,4-epoxy)cyclohexane-metadioxane (e.g., ERL-4234 from Union Carbide Corp.), bis(3,4-epoxycyclohexyl) adipate (e.g., ERL-4299 from Union Carbide Corp.), dipentene dioxide (e.g., ERL-4269 from Union Carbide Corp.), epoxidized polybutadiene (e.g., OXIRON 2001 from FMC Corp.), epoxy silanes (e.g., beta-(3,4-epoxycyclohexyl)ethyltrimethoxysilane and gamma-glycidoxypentyltrimethoxysilane, commercially available from Union Carbide), flame retardant epoxy resins (e.g., DER-542, a brominated bisphenol type epoxy resin available from Dow Chemical Co.), 1,4-butanediol diglycidyl ether (e.g., ARALDITE RD-2 from Ciba-Geigy), hydrogenated bisphenol A-epichlorohydrin based epoxy resins (e.g., EPONEX 1510 from Shell Chemical Co.), and polyglycidyl ether of phenolformaldehyde novolak (e.g., DEN-431, DEN-438, and DEN-439 from Dow Chemical Co.).

Useful photo-active curing agents are cationic and include aromatic iodonium complex salts, aromatic sulfonium complex salts, and metallocene salts, and are described in, for example, U.S. Patent No. 5,089,536 (Palazzotto). Peroxides and oxalate esters can be used with the metallocene salts to increase the cure speed, as described in U.S. Patent No. 5,252,694 (Willett). Useful commercially available photo-active curing agents include FX-512, an aromatic sulfonium complex salt (3M Company), CD-1010 an aromatic sulfonium complex salt from Sartomer, CD-1012, a diaryliodonium complex salt from Sartomer, an aromatic sulfonium complex salt (Union Carbide Corp.), UVI-6974, an aromatic sulfonium complex salt (Union Carbide Corp.), and IRGACURE 261, a metallocene complex salt (Ciba-Geigy). Photosensitizers may also be included, for example, to enhance the efficiency of the photo-active curing agent and/or to adjust the wavelength of photoactivity. Examples of photosensitizers include pyrene, fluoroanthrene, benzil, chrysene, p-terphenyl, acenaphthene, phenanthrene, biphenyl and camphorquinone.

A variety of thermally activated curing agents may also be incorporated into the compositions. For example, useful thermally activated curing agents include amine-, amide-, Lewis acid complex-, and anhydride-type materials and those which are preferred include dicyandiamide, imidazoles and polyamine salts. These are available from a variety of sources, e.g., Omicure™, available from Omicron Chemical, Ajicure™, available from Ajinomoto Chemical, and Curezol™, Amicure™ dicyandiamides (such as CG-1200 and CG-1400), and Ancamine™ (such as 2441 and 2337), available from Air Products.

In certain cases, it may be advantageous to add an accelerator to the composition, so that it will fully cure at a lower temperature, or will fully cure when exposed to heat for shorter periods. Imidazoles are useful, suitable examples of which include 2,4-diamino-6-(2'-methylimidazolyl)-ethyl-s-triazine isocyanurate; 2-phenyl-4-benzyl-5-hydroxymethylimidazole; and Ni-imidazole-phthalate.

Other useful blends for sealant layer 31 include epoxy-acrylate blends, such as those described, e.g., in Kitano et al., U.S. Patent No. 5,086,088. These blends are preferably the photopolymerized reaction product of a composition comprising (i) a prepolymeric (i.e., partially polymerized to a viscous syrup typically between about 100 and 10,000 centipoise) or monomeric syrup of an acrylic or methacrylic acid ester; (ii) optionally, a reinforcing comonomer; (iii) an epoxy resin; (iv) a photoinitiator; and (v) a thermally activated curing agent for the epoxy. Also useful is the thermally polymerized reaction product of a composition featuring (i) a prepolymeric (i.e., partially polymerized to a viscous syrup typically between about 100 and 10,000 centipoise) or monomeric syrup of an acrylic or methacrylic acid ester; (ii) optionally, a reinforcing comonomer; (iii) an epoxy resin; (iv) a thermal initiator; and (v) a photo-active curing agent for the epoxy. Suitable epoxy resins, and thermally activated curing agents include those described above. Examples of useful photoinitiators include quinones, benzophenones, triacylimidazoles, acylphosphine oxides, bisimidazoles, chloroalkyltriazines, benzoin ethers, benzil ketals, thioxanthenes, and acetophenone derivatives, and mixtures thereof. Examples of useful thermal initiators include organic peroxides and azo compounds. During use when the preferred sealants based on blends of epoxy-containing material and polyacrylate components softens, flows and fills gaps in the surface to be sealed, the epoxy resin and the polyacrylate component form a homogenous, single phase system, as evidenced by a lack of macroscopic phase separation to the unaided human eye.

The relative amounts of the different ingredients are selected to balance ultimate tensile strength and heat resistance, on the one hand, with flexibility and green strength build-up on the other hand. For example, increasing the amount of epoxy resin increases ultimate tensile strength and heat resistance, while decreasing flexibility and rate of green strength build-up. Conversely, increasing the amount of polyester or polyacrylate increases flexibility and rate of green strength build-up, while decreasing ultimate tensile strength and heat resistance.

In the case of epoxy-polyacrylate and epoxy-polyester blends, the compositions typically include from 0.01 to 95 parts by weight per 100 parts by weight total of the epoxy-containing material and, correspondingly, from 99.99 to 5 parts by weight of the polyester or polyacrylate component. More preferably, the compositions include from 0.1 to 80 parts by weight of the epoxy-containing material and, correspondingly, from 99.9 to 20 parts by weight of the polyester or polyacrylate component. Most preferably, the compositions include from 0.5 to 60 parts by weight of the epoxy-containing material, and, correspondingly, from 99.5 to 40 parts of the polyester or polyacrylate component.

Other melt-flowable thermosetting compositions useful for sealant layer 31 include urethane-based materials such as moisture-curable urethanes that may also be hot-melt compositions. Such compositions often comprise one or more polyisocyanates (e.g., diisocyanates such as 4,4'-diphenylmethylene diisocyanate, toluene diisocyanate, isophorone diisocyanate, or hexamethylene diisocyanate, including isocyanate derivatives of these materials), one or more polyhydroxy-functional materials (e.g., polyester or polyether polyols including polycaprolactones), optionally a catalyst for the moisture curing reaction (e.g., dibutyltin dilaurate), and optionally a variety of additives or adjuvants (e.g., fillers, colorants, beads, bubbles, fibers, plasticizers, tackifiers, flow control agents, thixotropes, adhesion promoters) that do not materially interfere with the moisture curing reaction.

A sealant layer may also be formed from a thermoplastic composition. Examples of suitable thermoplastic compositions include polyesters, thermoplastic elastomer block copolymers (e.g., styrene-butadiene- or styrene-isoprene-based block copolymers), thermoplastic elastomers, thermoplastic olefins, phenoxy resins, butyl rubbers, polyurethanes, silicones, and polyamides. Polyesters, block copolymers and polyurethanes are particularly preferred thermoplastics. Preferably, thermoplastic compositions used in the sealant layer are provided as homogenous, single phase materials that do not include a dispersed phase such as

cross-linked particles. Thermoplastic compositions selected for a sealant layer preferably display a softening temperature (as measured by a ring and ball softening test) that is greater than the service temperature for the ultimate construction into which the sealant-bearing article will be incorporated. The service temperature for the ultimate construction refers to the maximum temperature that the ultimate construction is expected to be exposed to under ordinary use conditions

Preferred compositions for a sealant layer are sealant compositions that resist flow, and thus substantially retain their shape, when heated to a temperature above the softening temperature of the sealant, and for thermosetting sealant compositions, a temperature that is less than (a) in the case of thermally activated curing agents, the thermal activation temperature of the curing agent or (b) in the case of photo-active curing agents, the thermal decomposition temperature of the curing agent, until subjected to pressure on the order of the pressure applied during installation as the tape-bearing substrate is pressed into contact with the other substrate. Under the influence of heat and applied pressure, these compositions undergo controlled flow to conform and functionally seal against uneven surfaces.

The softening temperature represents the minimum temperature at which the composition is sufficiently malleable such that it can be mounted to and held in place on a substrate. The softening temperature is a function of the particular sealant composition. In the case of crystalline or semi-crystalline component-containing sealing compositions, this generally corresponds to the melting temperature of this component. Typically, the upper temperature limit is on the order of about 200 °C.

Examples of compositions meeting these requirements include both thermoplastic and thermosettable materials. In the case of the latter, the compositions may incorporate one or more photo-active curing agents, thermally activated curing agents, or combinations thereof, with thermally activated curing agents being preferred.

Particular compositions meeting these requirements include the epoxy/polyester and epoxy/polyacrylate compositions described above, but particularly designed or formulated such that melt-flow does not occur under the influence of heat and gravity alone, but instead requires applied pressure as well. One useful formulation involves including one or more thixotropic agents into the composition in an effective amount; i.e., an amount necessary to achieve the desired rheological properties. In general, the total amount of thixotropic agents is no greater than about 20% by weight, based upon the total weight of the uncured sealant

composition, preferably no greater than about 10% by weight, more preferably no greater than about 5% by weight, and most preferably in the range of about 3-5% by weight.

Suitable thixotropic agents do not materially interfere with cure, in the case of thermosetting compositions, or otherwise cause degradation of the composition.

5 Representative examples of thixotropic agents include particulate fillers, beads (which may be of the glass, ceramic or polymeric type), bubbles (which may be of the glass, ceramic or polymeric type), and chopped fibers, as well as combinations thereof. Suitable particulate fillers include, e.g., hydrophobic and hydrophilic silica, carbon black, calcium carbonate, titania, clays such as bentonite, minerals such as felspar, and combinations thereof. Suitable
10 fibers include polymeric fibers (e.g., aromatic polyamide, polyethylene, polyester and polyimide fibers), glass fibers, graphite fibers, and ceramic fibers (e.g., boron fibers).

Other materials that can be incorporated into sealant layer 31 include, for example, stabilizers, antioxidants, plasticizers, tackifiers, flow control agents, adhesion promoters (e.g., silanes and titanates), colorants, and other fillers.

15 The tape includes an optional adhesive layer which may be disposed, for example, on either major surface of the core layer, either major surface of the sealant layer, or both. The adhesive layer, when present, is preferably in the form of a continuous layer. The width of the adhesive layer is application-dependent. In general, however, the width of adhesive layer is preferably no greater than the width of a core layer. In addition, both single and multi-
20 layer bonding compositions are envisioned.

In one embodiment, an adhesive layer is disposed between the core layer and the surface of the substrate to which the tape is affixed. In this case, the purpose of adhesive layer is to enhance adhesion between the substrate and the core layer. It may be integral with the tape, or it may be provided separately on the face of the substrate prior to affixing the tape
25 to the substrate. It is particularly useful when the substrate is glass.

The thickness of the adhesive layer is selected based upon the particular application for which the tape is to be used.

Suitable materials for the adhesive layer are tacky at the installation temperature. Both thermoplastic and thermosetting materials may be used. The adhesive layer is
30 ordinarily selected so as to have, as compared to the sealant layer, a different composition, thickness or both. The choice of a particular material for adhesive layer depends on the substrate to which the tape is affixed. For example, in the case of glass substrates,

thermosetting materials are preferred, whereas in the case of encapsulated glass substrates, in which a polymer encapsulates the peripheral edge of the glass, it may be preferred to use thermoplastic bonding materials.

5 Thermosetting materials may incorporate a photo-active curing agent (i.e., photo-curable materials) or a thermally activated curing agent (i.e., thermally curable materials). Preferably, the adhesive layer cures under conditions different from the conditions under which the sealant layer cures. For example, if both the sealant layer and the adhesive layer are photo-curable materials, the wavelength of radiation needed to initiate cure of the adhesive layer differs from that needed to initiate cure of the sealant layer. Similarly, if both
10 sealant and adhesive layers are thermally curable materials, they can cure at different temperatures.

Examples of suitable materials for the adhesive layer include epoxy/polyacrylate blends as described, e.g., in Kitano et al., U.S. Patent No. 5,086,088; epoxy/amorphous polyester blends; polyolefin adhesives (e.g., polyethylene, polypropylene, polyhexene,
15 polyoctene, and blends and copolymers thereof); ethylene-vinyl monomer (e.g., ethylene-vinyl acetate) copolymer adhesives; epoxy adhesives; silicone adhesives; silicone-acrylate adhesives; acrylic adhesives; rubber adhesives (e.g., butyl rubber); and adhesives based upon thermoplastic elastomer block copolymers (e.g., styrene-butadiene-styrene, styrene-isoprene-styrene, or styrene-ethylene-propylene-styrene block copolymers). These materials may be
20 provided in film or bulk form, and may be supplied as hot melt materials. Depending upon the substrate to which the adhesive layer will be adhered, the use of a primer may be advantageous in promoting adhesion. An example of a suitable commercially available material is 3M Company's Structural Bonding Tape No. 9214.

25 Other materials that can be incorporated into the adhesive layer include, for example, stabilizers, antioxidants, plasticizers, tackifiers, flow control agents, adhesion promoters (e.g., silanes and titanates), colorants, thixotropes, and other fillers.

Optional, temporary, protective liner 39, if included, protects the optional adhesive layer (if present) or core layer 32 from damage, actinic radiation exposure, and dirt or other contaminants until article 30 is intended for use and is typically removed shortly before
30 attaching article 30 to a substrate. Liner 39 may comprise a variety constructions, including those conventionally used to protect adhesive surfaces. For example, the liner may be in the form of a paper or polymeric web having a release material such as a polyolefin (e.g.,

polyethylene, polypropylene), silicone or fluorosilicone on a surface thereof that bears against the adhesive layer or the core layer. Liners that are slightly tacky can be used to protect non-tacky surfaces.

Manufacture

5 The multi-layer articles of the invention may be produced by laminating the various layers in any order suitable for the manufacturing process. In the simplest tape construction, an electrically conductive heating element is laminated to a layer of polymeric material. Lamination can be effected by laminating the heating element to a warm or hot extrudate as the polymeric material, e.g., a heat activated adhesive or sealant,
10 is extruded; it can be laminated to a sheet by warming the sheet and laminating; it can be laminated to a core that is inherently tacky or a core layer that has a pressure-sensitive adhesive coated on it; or it can be laminated to a pressure-sensitive adhesive. Additional layers can be bonded to the above described tape constructions at various times in the manufacturing process as desired. For example, a sealant layer is provided having a
15 heating element disposed on one surface, to which the heating element may or may not be bonded, and a core layer having a pressure-sensitive adhesive on both major surfaces is laminated by pressure to the mesh. The openings in the mesh provide sufficient adhesive to keep the tape laminated.

 Any of the layers may be made individually, and a laminated construction can be
20 formed at a later time using primers, adhesives, and the like under heat and/or pressure. However, it is not required that the layers of materials be joined. The individual layers can be provided separately to the end user, in for example a kit. The layers are then placed in a stack between two substrates and heat activated to join (bond and/or seal) the substrates. Alternatively, the unbonded layers of the tape construction can be help
25 together by one or more clips, staples, or the like. The clips may or may not be removed when the tape is used.

 Once the tape has been fabricated a release liner may optionally be laminated to protect the exposed surfaces of the sealant layer and/or the core layer or the adhesive layer (if provided). The tape may be converted into the desired final form by, for example,
30 slitting it to the desired width and winding it up into roll form and around a suitable plastic or paper core if needed. Alternatively, the tape can be slit or otherwise cut into discrete lengths or die cut into desired shapes.

Use

The above-described tapes can be used to establish a seal and/or a bond between a variety of substrates. Such substrates include glass, metal, painted metal, ceramic, wood, masonry, and plastic. Specific examples of how the tape may be used include bonding body
5 panels in vehicles such as cars, boats, trains, buses, and airplanes, attaching windows or windshields to window frames of vehicles and buildings, attaching signs to walls or windows, attaching carpeting or flooring to a subfloor, and bonding appliance panels. For the sake of simplicity, however, the sealing process will be described in the context of installing a glass windshield in a motor vehicle.

10 Assembly 230 (Fig. 13a) has a tape 180 having a tacky core layer, an electrically conductive heating element, and a sealant layer affixed to one face of glass windshield 232 through the core layer such that the tape substantially surrounds the perimeter of windshield 232 and adheres to the glass as the tape traverses the approximately ninety degree bends at the corners of the windshield. All of the bends form coplanar arcs, preferably with no
15 wrinkles or buckles on the tape. This arrangement avoids forming stress concentration points previously associated with the use of discontinuous spacers. The ends of the tape are preferably positioned at the bottom edge of the windshield, and preferably on one side to permit easy access to the ends of the tape when the windshield is placed into an automobile frame. The ends of the tape can be turned down 234 and connected to electrical leads 236
20 attached by wires 238 to power supply 240 capable of generating an electrical current. Alternatively, assembly 190 (Fig. 13b) has tape 250 which precisely fits the perimeter of windshield 232 so the tape ends are butted together but electrically isolated and electrical leads 242 are attached to the electrically conductive heating element at the butt joint. Lead wires 238 are connected to power supply 240 capable of generating an electrical current. In
25 another assembly 200 (Fig. 13c) the ends of tape 260 are joined into loop 264 such that the mesh from one end of the tape is attached to the mesh from the other end. In this use, loop 264 is positioned in an induction coil 264 so that when an electromagnetic field is generated by the coil, currents is induced in the mesh in the loop which causes an electrical current to flow through the tape.

30 If the core layer is not tacky at room temperature, it can be heated to bond the tape permanently to the glass, preferably without activating the sealant layer. Alternatively, an adhesive layer may be laminated to the core layer and the adhesive layer is affixed to the

glass surface. Because the sealant layer is not activated, the resulting tape-bearing windshield can be packed or racked in close proximity with other tape-bearing windshields without transferring sealant to a neighboring windshield. The tape also prevents the windshields from bumping into each other which eliminates costly packaging materials that space adjacent racked or packed windshields from each other (e.g., polymeric foam or cellulosic spacers) and which may require separate disposal or recycling.

The next step is to heat the sealant by generating an electrical current through the electrically conductive heating element. Preferably, the sealant is heated to the point where it softens but does not flow. As represented by Fig. 16, windshield 184 bearing tape 187 containing the heated, softened sealant is then installed in the frame of a motor vehicle 181. Lead wires 183 are positioned at a lower side 185 of vehicle 181 to facilitate access to the wires. It is also possible to heat the sealant after installing it in the motor vehicle frame to soften the sealant. During installation, pressure is applied that causes the softened sealant to flow and "self level" with the core layer relative to the uneven surface of the vehicle. The sealant flows away from high spots and fills in recessed areas such as spot welds and cavities, creating an effective seal. In severely distorted metal areas, the core layer compresses upon itself and may be permanently deformed in the process of creating a seal with the uneven surface.

Although it is preferable to include the sealant layer, core layer, and adhesive layer in the form of a single integral tape, it is also possible to apply these materials separately, or in various combinations with each other, to the glass surface. For example, it is possible to apply a tape having electrically conductive heating element, the core layer and the adhesive layer to the glass surface, followed by application of a separate sealant layer.

Alternatively, the adhesive layer may be provided in the form of a primer applied to the glass surface, after a tape containing the sealant layer, the core layer, and an electrically conductive heating element is affixed to the primed surface. In another alternative method of use, the individual layers, e.g., the sealant layer, the core layer, and the electrically conductive heating element, may be stacked on each other unbonded or partially bonded, and then bonded by applying a current to the electrically conductive heating element.

In yet another alternative, the core layer is bonded to the glass surface either with or without the heating element bonded to it. A sealant, which may be for example, a pumpable paste or a sealant tape or rope, is disposed on the second surface. If the electrically

conductive heating element is not already attached to the core layer, it can be attached to the core layer or bent and laid over the sealant. The core layer, attached to the glass is then placed over the electrically conductive heating element. The heating element is then connected to a power supply and current applied to bond the substrates.

5 It is noted that it is not particularly important when the lead wires are attached to the ends of the electrically conductive heating element during the process. The lead wires may be attached to heating element in the tape on the windshield, or they may be attached after the windshield has been positioned in the car window frame for installation, or in the case where the heating element is separately positioned into from, they may attached after the heating
10 element is put in place but before the windshield is put in place.

Although in the case of substrates such as windshields it is preferable to apply the tape to a face of the substrate, it is also possible to apply the tape around the edge of a substrate that the tape substantially encircles the substrate. Such constructions may be useful, e.g., in architectural applications for bonding the substrate within a groove such as a window
15 frame.

The invention can also be used to facilitate removal of one substrate from a second substrate in a bonded assembly having a tape with at least one polymeric layer and an electrically conductive heating element. The electrically conductive heating element may or may not have been used to bond the assembly. The ends of the electrically conductive
20 heating element are exposed if they aren't exposed already, and connected to a power source. Sufficient current is applied to soften the polymeric material to permit easier removal of the first substrate.

A power supply is used to provide power to the electrically conductive heating element in the tape. The power supply is preferably capable of supplying 0 to 60 volts of direct current (VDC) at 0-40 amps. In practice, the amount of power needed to bond the
25 tape the a substrate is determined and the power supply is set to provide the desired VDC output. The amount of voltage and the time the output voltage is applied to the electrically conductive heating element is controlled as determined by the length and width of the tape to be bonded.

30 The power supply typically has of three major components - an input supply circuitry, an operator control, and an output circuitry. The input supply circuitry is a 60

VDC power supply capable of supplying 40 Amps of current. This input supply could be a linear or switching DC supply powered by an AC voltage or a set of batteries.

This controller is a single turn knob that adjusts the resistive potentiometer (pot). This pot alters the voltage to the output circuitry chip on the output control board, which in turn changes the output voltage to the electrically conductive heating element.

The 0-60 VDC output circuitry has a Pulse Width Modulation (PWM) voltage. A PWM voltage is a percentage of cycled DC voltage from the input power supply. Preferably, the PWM control causes the 60 VDC input supply to oscillate from 0 VDC to 60 VDC at a given percentage. The final output voltage to the electrically conductive heating element is a quasi-average of the percent of 0 VDC and 60 VDC time. For example, if the control pot is set to provide 20 VDC output to heating element, the PWM circuitry outputs a 60 VDC pulse for approximately 33% of the time and shuts the output voltage off (0 VDC) for approximately 67% of the time. These pulses occur at a 22.5 kHz rate and percentages will vary due to differences in the type of heating element and the size of the tape used to bond and/or the substrates.

The invention will now be described further by way of the following non-limiting examples.

Test Methods

Test Method A – Coplanar Arc Bend Test A1, A2 and A3

These tests are used to determine the ability of an article having an electrically conductive heating element to bend into a coplanar arc (Test A1), a coplanar arc around a 10 cm radius circle (Test A2), and a coplanar arc around a 4 cm radius circle (Test A3).

An article that is capable of passing test A3 would be capable of passing both test A1 and A2 since test A3 is more stringent than A2 and test A2 is more stringent than A1. Likewise an article that is capable of passing test A2 would be capable of passing test A1.

Coplanar Arc Bend Test A1

If the article to be tested is not tacky, it is first attached to a conformable pressure-sensitive adhesive tape (3M™ 4941 Acrylic Foam Tape available from Minnesota Mining & Manufacturing Co.) such that the test article is coextensive with the pressure-sensitive

adhesive tape. If the article is very stiff at room temperature, a hot air gun can be blown on the article/optional tape combination to soften it before or during application. The article/optional tape combination is placed on a flat surface and a first plane is located on the flat surface; a second plane is located substantially on the top surface of the article/optional tape combination and parallel to the first plane. The article/optional tape combination is then bent laterally by hand while simultaneously attaching it to the flat surface such that the lateral edges of the article/optional tape combination form concentric arcs and the outside arc has a length that is at least about 1% greater than the inside arc. The bent article/optional tape combination should adhere to the surface. If the article/optional tape combination adheres smoothly to the surface, substantially all of the article/optional tape lies between the two planes. If small wrinkles or buckles exist, the amount of article/optional tape combination that lies outside of the two planes due to wrinkling and buckling is visually estimated. The amount of article/optional tape combination lying outside of the second plane should be less than about 25% in order to pass this test. Preferably it should be less than about 20%, and more preferably, less than about 10%, and most preferably 0 %.

Test results are reported in the amount of tape lying outside of the second plane in percent for the arc. A rating of 0 percent indicates that substantially all of the article/optional tape combination remained between the two planes after bending while a rating of 90 percent indicates extremely severe buckling or lifting of the article/optional tape combination.

Coplanar Arc Bend Test A2

A test substrate (a sheet of 6 mm thick foam board having a smooth, flat surface) is prepared by drawing a circle having a radius of 10 cm on it. The article/optional tape combination is prepared in the manner described in Test A1 above. The article/optional tape combination is placed on the foam board and the first and second planes are identified such that the foam board is the first plane and the second plane is identified in a similar manner as in Test A1. The article/optional tape combination is then bent laterally around the circle for at least 90 degrees such that the inside edge of the article/optional tape combination is aligned to an arc on the circumference of the drawn circle and the outside

edge is substantially aligned with the arc of a concentric circle having a radius of approximately 10 cm plus the width of the article/optional tape combination.

5 The amount of article/optional tape combination lying outside of the second plane should be less than about 25% in order to pass this test. Preferably less than about 20%, and more preferably, less than about 10%, and most preferably 0 percent.

10 Test results are reported in the amount of tape lying outside of the second plane in percent for an arc of a circle having a 10 cm radius. A rating of 0 percent indicates that substantially all of the tape remained between the two planes after bending while a rating of 90 percent indicates extremely severe buckling or lifting of the article/optional tape combination.

Coplanar Arc Bend Test A3

An article/optional tape combination is tested in the manner described in Test A2 except the circle drawn has a 4 cm radius.

15 The amount of article/optional tape combination lying outside of the second plane should be less than about 25% in order to pass this test. Preferably less than about 20%, and more preferably less than about 10%, and most preferably 0 percent.

20 Test results are reported in the amount of tape lying outside of the second plane in percent for an arc of a circle having a 4 cm radius. A rating of 0 percent indicates that substantially all of the tape remained between the two planes after bending while a rating of 90 percent indicates severe buckling or lifting of the article/optional tape combination.

Test Method B-Coplanar Arc Bend Tests B1, B2, and B3 for a Heating Element

25 These tests are used to determine the ability of an electrically conductive heating element to bend into a coplanar arc (Test B1), a coplanar arc around a 10 cm radius circle (Test B2), and a coplanar arc around a 4 cm radius circle (Test B3).

30 A heating element that is capable of passing test B3 would be capable of passing both test B1 and B2 since test B3 is more stringent than B2 and test B2 is more stringent than B1. Likewise a heating element that is capable of passing test B2 would be capable of passing test B1.

Coplanar Arc Bend Test B1

This test is used to determine a preferred heating element. A tape/heating element combination is prepared by attaching the heating element to a pressure-sensitive adhesive tape (3M™4941). The tape/heating element combination is positioned between two planes and attached to a surface as described in Test Method A1. The heating element on the tape is then examined visually. Any buckling or lifting of the heating element is less than about 7 times the thickness of the heating element as measured from the surface of the tape in order to pass this test. Heating elements that are loftier in nature, such as knit and nonwoven meshes, are considered to pass if any protruding wires of the heating element can be tacked down by pressing the wires onto the adhesive tape.

For example, if the heating element were 0.005 inch (0.127 mm) thick, any bending or warping of the heating element out of the plane of the mesh would be no greater than 0.035 inch (0.9 mm). Preferably the heating element remains co-planar after being bent without buckling or lifting more than about 5 times the thickness of the heating element, more preferably without buckling or lifting more than about 3 times the thickness of the heating element, and most preferably with no buckling. Additionally, when the heating element is a mesh, the mesh, after it has been bent around the circle, preferably does not neck down in width more than about 10 %, and more preferably it does not neck down in width more than about 5%.

Coplanar Arc Bend Test B2

This test is used to determine a preferred heating element. A tape/heating element combination is prepared by attaching the heating element to a pressure-sensitive adhesive tape (3M™4941). The tape/heating element combination is positioned between two planes and attached to a surface as described in Test Method A2. The heating element on the tape is then examined visually. Any buckling, kinking, or overlapping of the heating element is typically less than 7 times the thickness of the heating element as measured from the surface of the tape. For example, if the heating element were 0.005 inch (0.127 mm) thick, any bending or warping of the heating element out of the plane of the mesh would be no greater than 0.035 inch (0.9 mm). Preferably the heating element remains co-planar after being bent without buckling more than 5 times the thickness of the heating element, and most preferably without buckling more than 3 times the thickness of the

heating element. Heating elements that are three dimensional in nature, such as knit and nonwoven meshes, are suitable if the protruding wires of the mesh can be tacked down by pressing the wires onto the adhesive tape.

5 Additionally, when the heating element is a mesh, the mesh, after it has been bent around the circle, preferably does not neck down in width more than about 10 %, and more preferably it does not neck down in width more than about 5%.

Coplanar Arc Bend Test B3

10 This test is used to determine a preferred heating element. A tape/heating element combination is prepared by attaching the heating element to a pressure-sensitive adhesive tape (3M™4941). The tape/heating element combination is positioned between two planes and attached to a surface as described in Test Method A3. The heating element on the tape is then examined visually. Any buckling, kinking, or overlapping of the heating element is typically less than 7 times the thickness of the heating element as measured
15 from the surface of the tape. For example, if the heating element were 0.005 inch (0.127 mm) thick, any bending or warping of the heating element out of the plane of the mesh would be no greater than 0.035 inch (0.9 mm). Preferably the heating element remains coplanar after being bent without buckling more than 5 times the thickness of the heating element, and most preferably without buckling more than 3 times the thickness of the
20 heating element. Heating elements that are three dimensional in nature, such as knit and nonwoven meshes, are suitable if the protruding wires of the mesh can be tacked down by pressing the wires onto the adhesive tape

25 Additionally, when the heating element is a mesh, the mesh, after it has been bent around the circle, preferably does not neck down in width more than about 10 %, and more preferably it does not neck down in width more than about 5%.

Test Method C-Elongation for Preferred Meshes

This test is a used to determine preferred meshes for the practice of the invention. A strip of mesh measuring 0.635 cm is clamped into the jaws of an Instron™Tensile Tester
30 with a jaw separation of 5.08 cm. The jaws are separated at a speed of 20 inches (50.8 cm) per minute. The elongation is recorded at 1 pound of force (4.45 Newtons) and at 2 pounds of force (8.90 Newtons). Preferred meshes should have an elongation of greater

than 0.5% at 1 pound of force, and greater than 1% at 2 pounds of force. More preferred meshes have and elongation at 1-pound of greater than 3% and at 2 pounds of greater than 5%.

5 DEFINITION OF MESHES USED IN EXAMPLES

All meshes were obtained from Delker Corp. unless otherwise indicated.

1. 4 Ni 6-100 - A nickel foil having an original thickness of 0.004 inch, a strand width of 0.006 inch, and an LWD of 0.100 inch
- 10 2. 5 Ni 10-125 - A nickel foil having an original thickness of 0.005 inch, a strand width of 0.010 inch, and an LWD of 0.125 inch
3. 5 Cu 14-189 - A copper foil having an original thickness of 0.005 inch, a strand width of 0.014 inch, and an LWD of 0.189 inch
4. 5 Cu 20-284 - A copper foil having an original thickness of 0.005 inch, a strand width of 0.020 inch, and an LWD of 0.284 inch
- 15 5. 5 Ni 5-050P Nickel - A pulled nickel foil having an original thickness of 0.005 inch, a strand width of 0.005 inch, and an LWD of 0.050 inch
6. An5 Fe 5-050 Ni plated - A nickel plated annealed steel foil having an original thickness odddf 0.005 inch, a strand width of 0.0005 inch, and an LWD of 0.125 inch
7. 5Fe 7-125 Ni plated - A nickel plated steel foil having an original thickness of 0.005
20 inch, a strand width of 0.007 inch, and an LWD of 0.125 inch
8. 7Ni7-4/3 DB Nickel (available from Exmet Corp.) A nickel foil having an original thickness of 0.007 inch, a strand width of 0.007 inch, and an LWD of 0.100 inch
9. 3 Ni 5-125 A nickel foil having an original thickness of 0.008 inch, a strand width of 0.012 inch, and an LWD of 0.125 inch
- 25 10. 5 Ni 7-125 A nickel foil having an original thickness of 0.005 inch, a strand width of 0.007 inch, and an LWD of 0.125 inch

Examples 1-4

30 A tape 80 (Fig. 8) for each example was formed by laminating 1.27 cm wide expanded metal foil 84, i.e, mesh, to a core layer 82 (3M™4941 pressure-sensitive adhesive tape, having an acrylic adhesive on each major surface of an acrylic foam, available from Minnesota Mining & Manufacturing Co.) having a width of 1.27 cm and a

thickness of 1.2 mm on release liner 86. The specific mesh used for each example is shown in Table 1. The resulting tapes were bent into coplanar arcs as described in Test Method A, and the amount of tape protruding above the second plane are shown in Table 1.

5

Table 1				
Ex	Mesh Type	Coplanar Arc-%	10 cm radius arc-%	4 cm radius arc-%
1	4 Ni 6-100	0	0	0
2	5 Ni 10-125	0	0	0
3	5 Cu 4-189	0	0	0
4	5 Cu 20-284	0	0	0

Examples 5-8

An epoxy resin mixture was prepared by mixing 50 parts of an epoxy resin (Epon™ 828) with 50 parts of a second epoxy resin (Epon™ 1001) in a paint can heating in an oven at 100°C for 2 hours. A sealant composition was prepared by mixing 85 parts of polyester (Dynapol™1402), 15 parts of the epoxy resin mixture, and 6 parts of an amine epoxy curative (Ancamine 2441 available from Air Products and Chemicals, Inc.) and feeding a twin screw extruder. In a downstream port, 2.5 parts of silica (Aerosil R972 available from Degussa Corp.) were added in. The barrel temperatures were set at 80°C. The extrudate was cast onto a release liner in a chilled roller nip and calendered into a sheet of sealant having a thickness of 2 mm. A sheet of electrically conductive mesh (type for each example shown in Table 2) was also fed into the chilled roller nip and laminated to the surface of the sealant. Meshes for Examples 5-7 were obtained from Delker Corp. and the mesh for Example 8 was obtained from Exmet Corp.). After cooling, the mesh surface of the sealant was laminated to a tacky core layer (3M 4941 Adhesive Tape available from Minnesota Mining & Manufacturing Co.).

The sealant and core layer were removed from approximately 1.27 cm on each end of a tape measuring 12.7 cm by 1.27 cm wide to expose the mesh. The tape was placed between two glass plates that had been cleaned with 3M™ Glass Cleaner (available from Minnesota Mining & Manufacturing Co.). Then the exposed ends of the mesh were connected to copper alligator clips which were soldered to copper wires connected to a constant voltage power source (Hewlett Packard 6032A 0-60V / 0-60A / 1000W available from Hewlett Packard). The current (Amps), shown in Table 2, was applied to the system

for 10 minutes at the voltage (Volts) setting shown. The resistance in Ohms was calculated as shown. The samples were cooled for 5 minutes and then disconnected from the power source. All of the samples showed that the sealant had flowed sufficiently to wet out on the surface. Additionally, Examples 7 and 8 each formed a tight bond between the glass plates.

Table 2				
Ex	Mesh Type	Voltage-Volts	Current-Amps	Resistance-Ohms
5	5Ni5-050P Nickel	2.05	17.31	0.118
6	An5Fe5-050 Ni plated steel	1.80	17.34	0.104
7	5Fe7-125 Nickel plated steel	2.01	10.41	0.193
8	7Ni7-4/3 DB Nickel	1.31	19.70	0.066

Example 9

A 111.8 cm x 1.27 cm tape was prepared according to the procedure of Example 5 except that the mesh used was 3Ni5-125 available from Delker Corp. The sealant and core layer were removed from about 1.27 cm from each end of the tape. The tape was positioned between two cleaned glass plates in a continuous strip so that it had four curves which approximated a 90 degree arc on a circle having a radius of 4 cm. Alligator clips were attached to the mesh and connected to the power source. Then current was applied (30 volts and 5.9 amps) to the system for 10 minutes followed by a 5 minute cooling cycle. The final resistance was calculated to be 5.08 ohms.

The assembly was disconnected from the alligator clips and cooled completely. The glass plates are positioned upright and tap water at ambient temperature was poured into the cavity formed by the two glass plates and the tape such that the cavity was about half full of water. No leaking of water was observed in the test assembly after one hour, thus showing that a tape of the invention can form a water tight seal on glass.

Example 10

Assembly (Fig. 14) was prepared by cleaning substrate 218 with 3M Glass Cleaner. Substrate 218 was an automotive windshield. A 0.635 cm wide tape, prepared according to the procedure of Example 5 except that the mesh 212 was 5Ni7-125. Tape 180 having adhesive layer 216, core layer 214, electrically conductive heating mesh 212, and sealant layer 210, was laminated around the inside periphery of the windshield substrate 218 by pressing the tape onto the glass and bending it around the corners. The

tape formed a series of coplanar arcs around the windshield. The sealant, core, and adhesive were removed from the ends of the tape, exposing the mesh. The ends of the tape were brought into close proximity to each other at the bottom of the windshield 234 and turned downward. The windshield was positioned onto an automobile frame (not shown). Each end of the mesh was connected to an electrical lead 236 which was soldered to lead wire 238 connected to power supply 240. Then 50 volts of power were applied for two minutes, then the voltage was increased to 60 volts (8.3 amps) for 8 minutes. The temperature between the glass and sealant was measured to be 250°F. The sealant had sufficient flow to seal and securely bond the windshield to the frame after cooling to ambient temperature.

Example 11

A 2 mm thick sealant layer 68 (Fig. 7) laminated to a metal mesh 24 was prepared according to the procedure of Example 5. A second 2 mm thick sealant layer 66 was laminated to the first sealant layer by warming the surfaces of the sealant to about 80°C and pressing the surfaces together. Then a first core layer 61 having a pressure-sensitive adhesive layer (not shown) on each major surface of core 61 was laminated to mesh 24 by hand. A second core layer 63, also having a pressure-sensitive adhesive layer (not shown) on each major surface of core 63 was laminated to core layer 61. Core layer 63 also included release liner 39. Core layers 63 were both 3 mm thick acrylic foam tape (3M™ 4959 available from 3M Co.) The resulting tape 70 was bent into a coplanar arc, as well as a coplanar arc of a 10 cm radius circle and a 4 cm radius circle with all of the tape lying between the first and second planes as described in Test Method A.

Example 12

A sheet was prepared by laminating a conductive heating element (5-Ni-10-125 Mesh) to a 2mm thick sheet of the sealant material of Example 5. A tape was prepared by cutting a strip measuring 48 cm by 1.27 cm and removing the sealant from 1.27 cm on each end of the strip exposing heating element. Then two layers of 1.2 mm thick acrylic foam tape having an acrylic pressure-sensitive adhesive on each major surface (3M 4941 available from Minnesota Mining & Manufacturing Co.) to the heating element. The adhesive side of the tape was then laminated to a cleaned glass plate leaving the sealant

exposed. A second sealant material (Macromelt 6240 available from Henkel Corp.) was extruded into a 0.5 mm thick sheet. A strip of the second sealant measuring 45.7 cm by 1.27 cm was placed over the exposed sealant surface. Four metal strips measuring 7.6 cm by 10.2 cm were primed with a primer which was a 0.5% by weight solution of glycidoxypyltrimethoxy silane in a 50/50 mixture of isopropanol and water and dried. The coupons were then laid over the second sealant layer with the 10.2 cm dimension running lengthwise along the tape and with the primer side against the sealant. Alligator clips, connected to a power source, were attached to the exposed ends of the heating element. The power source was turned on to a voltage of 9.2 volts for 10 minutes, and then turned off. The entire assembly was cooled to room temperature before removing the alligator clips. Both sealant layers had melted and flowed out and adhered to the metal coupons, while the acrylic foam core maintained the desired spacing between the metal and the glass.

Example 13

A 0.5 mm diameter copper wire was arranged along the length of a threaded rod and another length of the same type of wire was wound around the threaded rod and the first wire in a spiral fashion, using the threads of the threaded rod as a guide. Then each point where the spiral winding intersected with the linear wire was soldered. The soldered assembly was removed from the threaded rod, and each turn of the spiral winding was cut at a point opposite the solder joint. The curved appendages were straightened and trimmed to form an electrically conductive heating element having a single conductor 111 (Fig. 11a) with linear appendages 113. The width of the heating element was about 12.7 mm.

A 10.5 mm long section the electrically conductive heating element was placed in a rectangular shaped teflon mold that was about 12.7 mm wide and 3.2 mm deep. The ends of the electrically conductive heating element were covered with silicone rubber to form dams at each end. A polyester resin (Dynapol S1402) was heated until it was molten and then poured into the mold to a depth of about 3.2 mm. After cooling and hardening, the resulting article in tape form measuring 12.7 mm wide, 3.2 mm thick, and 10.5 mm long was removed from the mold. The tape, when heated to about 100F can be bent into a coplanar arc. The protruding ends of the electrically conductive heating element were

attached to a power source and heated. The heating element effectively melted the polyester resin.

Examples 14 – 20 and Comparative Examples C1-C2

5 Meshes were cut to a width of 0.635 cm and tested for elongation as described in Test Method C. The elongation in percent of the original length was measured at 1 pound (4.45 N) and at 2 pounds (8.90 N). The tape construction of Example 14 was a tape construction having a sealant layer, a core layer, and an expanded metal mesh (5Ni10-125). Example 15 was a loosely knit copper wire mesh. Examples 16-19 were meshes
10 formed by piercing and expanding metal foils. Example 20 was the woven welded screen of Example 22 cut on a bias such that there were essentially no straight wires running longitudinally. Example 21 was a woven welded metal screen having a spacing of 0.635 cm with straight wires running longitudinally. The tested mesh included 2 wires. Example 22 was a woven welded metal mesh having a spacing of 0.318 cm with wires
15 running longitudinally.

Ex	Sample Description	% Elongation-4.45 N	% Elongation-8.90 N
14	Tape Construction	4.2	8.3
15	Loose knit copper wire mesh	13.6	27.5
16	4Ni6-100 expanded metal	26.4	52.8
17	5Ni7-125 expanded metal	7.15	14.3
18	5Cu20-284 expanded metal	74	148
19	5Cu14-189 expanded metal	80.8	161.7
20	C2 cut on a bias	10.3	20.5
C1	Welded metal screen	0.42	0.85
C2	Woven welded metal screen	0.25	0.49

Example

20 A sealant layer was prepared by extruding a thermoplastic polyester type polyurethane (Estane 58213 available from B.F. Goodrich) into a 2 mm thick sheet. The sealant was then laminated to an acrylic pressure-sensitive adhesive foam tape (3M™4941) with an electrically conductive mesh (4Ni6-100) between the foam tape and the sealant layer. A 10.2 cm by 1.27 cm tape was cut from the sheet and the foam side was attached to a cleaned glass plate with the sealant layer exposed. The ends of the mesh were
25 exposed and connected to a power supply set at 2.2 volts. A second cleaned glass plate

was placed over the sealant layer. Power was applied for 4 minutes, during which time the sealant softened to seal the glass plates together. The power supply was disconnected and the sample was cooled for 30 minutes. At this point, the plates could not be separated by hand. The ends of the mesh were then reconnected to the power supply, set at 2.2 volts, 5 for 2 minutes. The power source was disconnected and the sealant had softened sufficiently to allow the glass plates to separate at the interface between the heating element and the foam adhesive.

10 Various modifications and alterations of this invention will become apparent to those skilled in the art without departing from the scope and spirit of this invention, and it should be understood that this invention is not to be unduly limited to illustrative embodiments set forth herein.

We claim:

1. An article comprising:

a laminate, the laminate comprising:

(a) a heating element selected from the group consisting of:

5 (i) an electrically conductive mesh;

(ii) a component comprising a continuous electrically conductive path, the path having a plurality of thermally conductive appendages attached to the continuous electrically conductive path in such a manner that heat generated in the continuous path by electricity flowing therethrough can be thermally conducted through the appendages;

10 (b) a polymeric material;

wherein the article is capable of bending according to Coplanar Arc Bend Test A1.

2. The article of claim 1 wherein the polymeric material comprises a heat activatable material selected from the group consisting of thermosettable materials, thermoplastic materials, and mixtures thereof, wherein the heat activatable material is in thermal contact with the heating element, such that if the heat activatable material is not in direct contact with the heating element it is separated from the heating element by a material that is thermally conductive to a sufficient degree to allow activation of the heat activatable material, and wherein the heating element is capable upon having an electrical current flow therethrough of causing at least a portion of the heat activatable material to soften, melt, and/or cure.

3. The article of claim 1 wherein the polymeric material comprises a conformable, compressible melt flow-resistant thermoset core.

4. The article of claim 1 wherein the heating element comprises a mesh.

5. The article of claim 1 wherein the heat activatable material is selected from the group consisting of sealants and adhesives.

6. An assembly comprising:

(a) a first substrate;

(b) the article of claim 1, 2, or 3 joined to the first substrate.

7. The assembly of claim 6 wherein the assembly further comprises a second substrate and wherein the first substrate and second substrate are joined together via the article.

5

8. The assembly of claim 6 wherein the first substrate is selected from the group consisting of vehicle glazing, architectural glazing, computer screens, television screens, vehicle body panels, carpeting, and flooring.

10

9. The assembly of claim 6 wherein the first substrate comprises a windshield.

10. A method comprising the steps of:

(a) providing a first substrate;

(b) providing a stack of materials in contact with the first substrate,

15

wherein the stack of materials comprises:

(i) a heating element selected from the group consisting of:

(A) an electrically conductive mesh, wherein the mesh is capable of passing Coplanar Arc Bend Test B1;

20

(B) a component comprising a continuous electrically conductive path, the path having a plurality of thermally conductive appendages attached to the continuous electrically conductive path in such a manner that heat generated in the continuous path by electricity flowing therethrough can be thermally conducted through the appendages, and wherein the component is capable of passing Coplanar Arc Bend Test B1 ;

25

(ii) a heat activatable material selected from the group consisting of thermosettable materials, thermoplastic materials, and mixtures thereof, wherein the heat activatable material is in thermal contact with the heating element, such that if the heat activatable material is not in direct contact with the heating element it is separated from the heating element by a material that is thermally conductive to a sufficient degree to allow activation of at least a portion of the heat activatable material, and wherein the heating element is

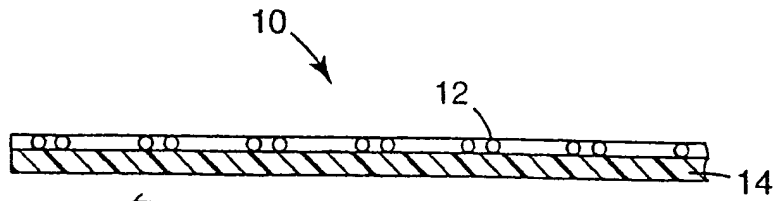
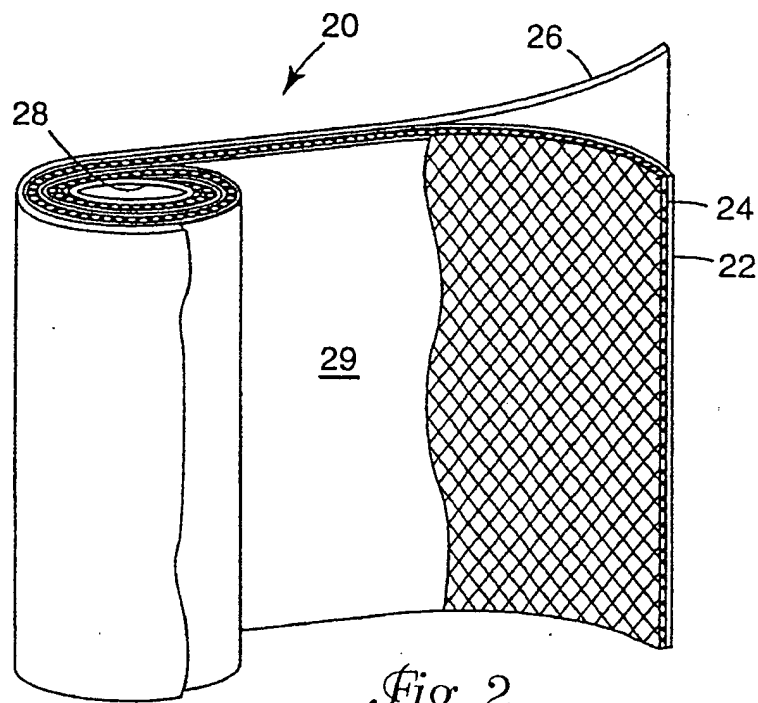
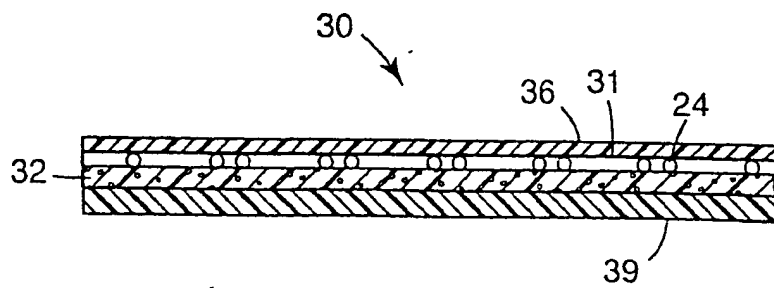
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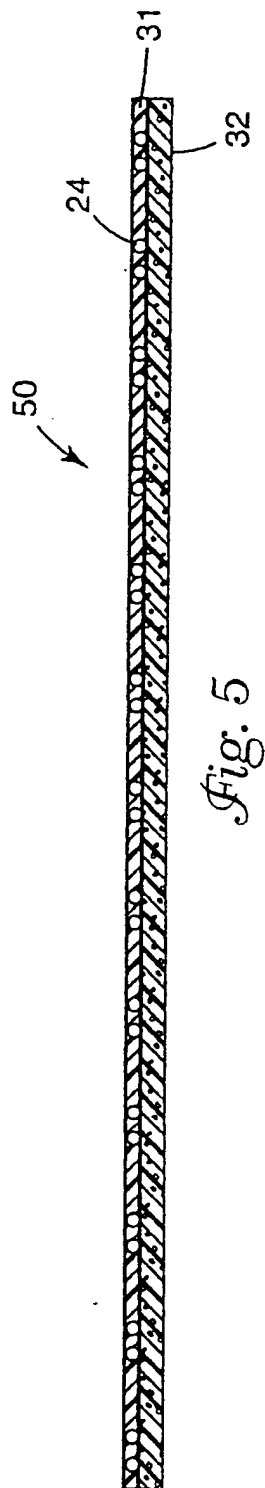
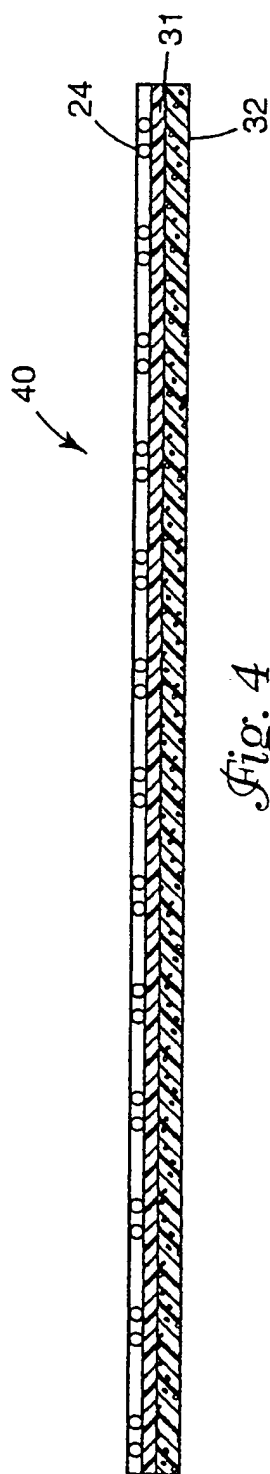
capable upon having an electrical current flow therethrough of causing at least a portion of the heat activatable material to soften, melt, and/or cure;

- (c) placing a second substrate against an exposed surface of the stack; wherein alternatively the stack is simultaneously provided between the first and second substrate;
- (d) causing an electrical current to flow through the heating element during one or more of the following: step (b), step (c), after step (c); in a manner to cause the heat activatable material to soften, melt, and/or cure such that the first substrate is ultimately joined to the second substrate through the stack in order to form an assembly.

11. An assembly comprising:

- (a) a first substrate;
- (b) a second substrate;
- (c) a laminate joining the first and second substrate together wherein the laminate comprises a
- (i) a heating element selected from the group consisting of:
- (A) an electrically conductive mesh, wherein the mesh is capable of passing Coplanar Arc Bend Test B1;
- (B) a component comprising a continuous electrically conductive path, the path having a plurality of thermally conductive appendages attached to the continuous electrically conductive path in such a manner that heat generated in the continuous path by electricity flowing therethrough can be thermally conducted through the appendages, and wherein the component is capable of passing Coplanar Arc Bend Test B1;
- (ii) a polymeric material.

*Fig. 1**Fig. 2**Fig. 3*



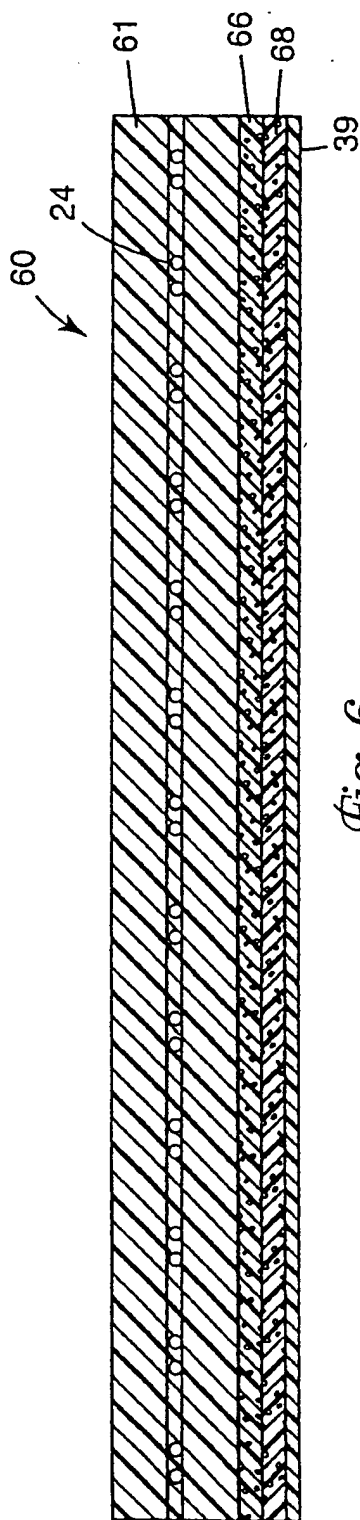


Fig. 6

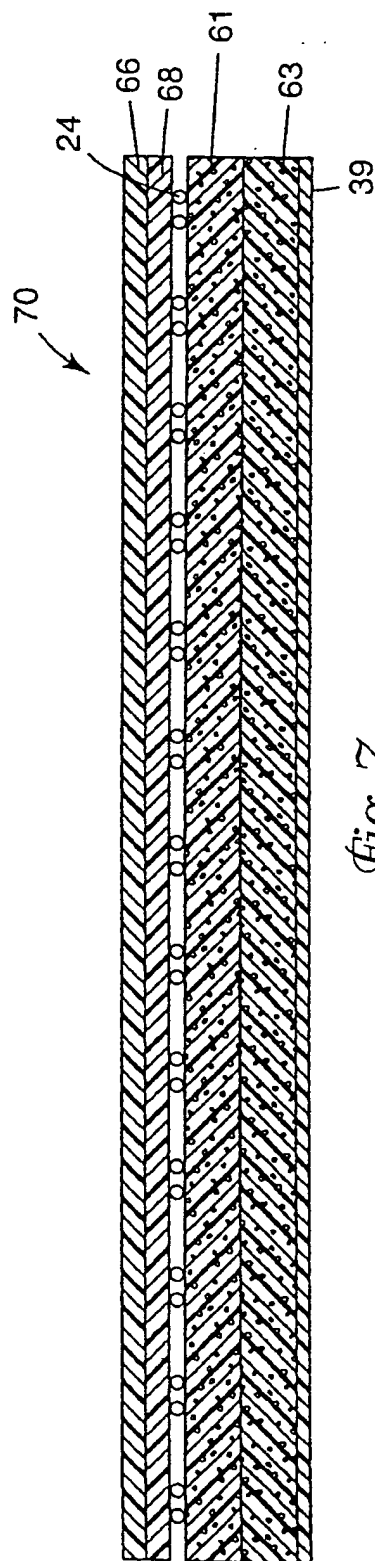


Fig. 7

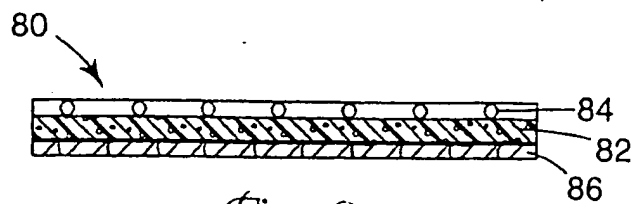


Fig. 8

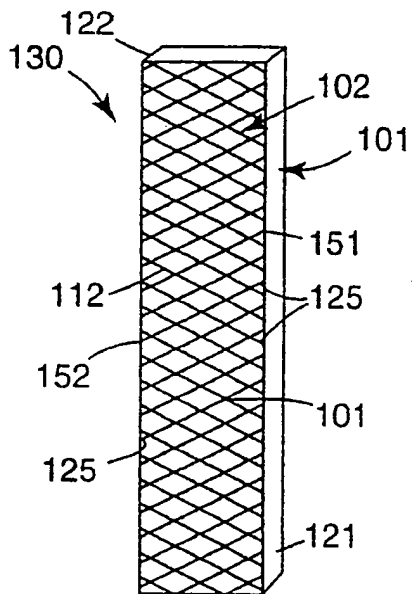


Fig. 9

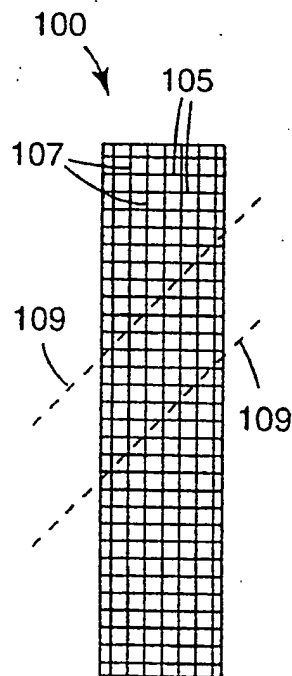


Fig. 10

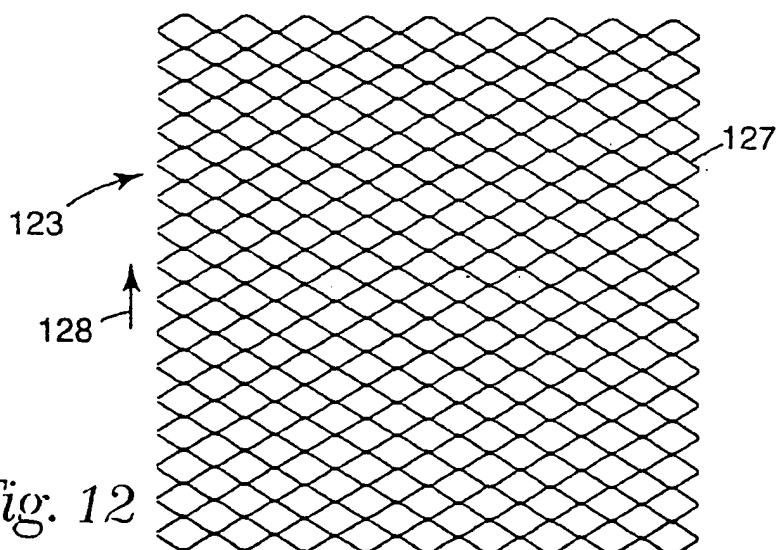


Fig. 12

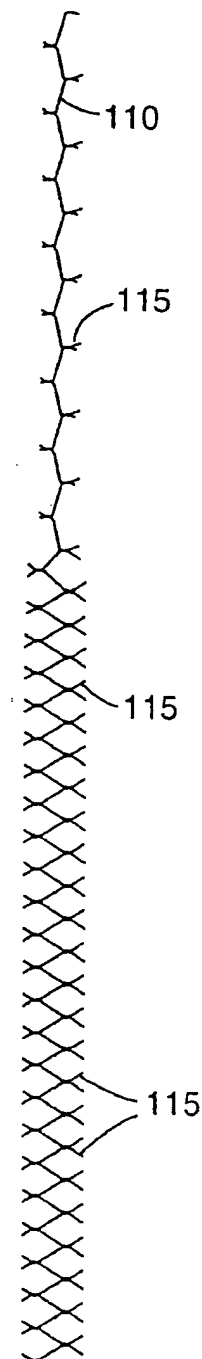
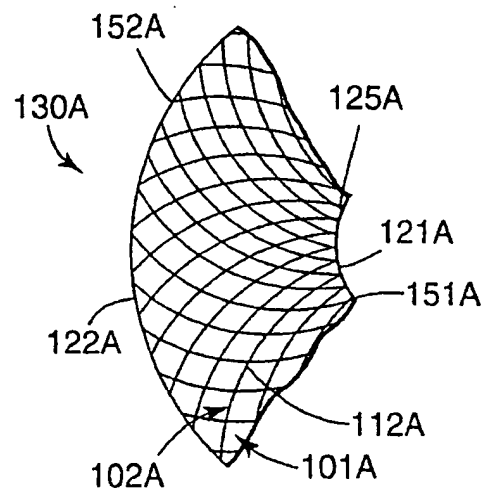
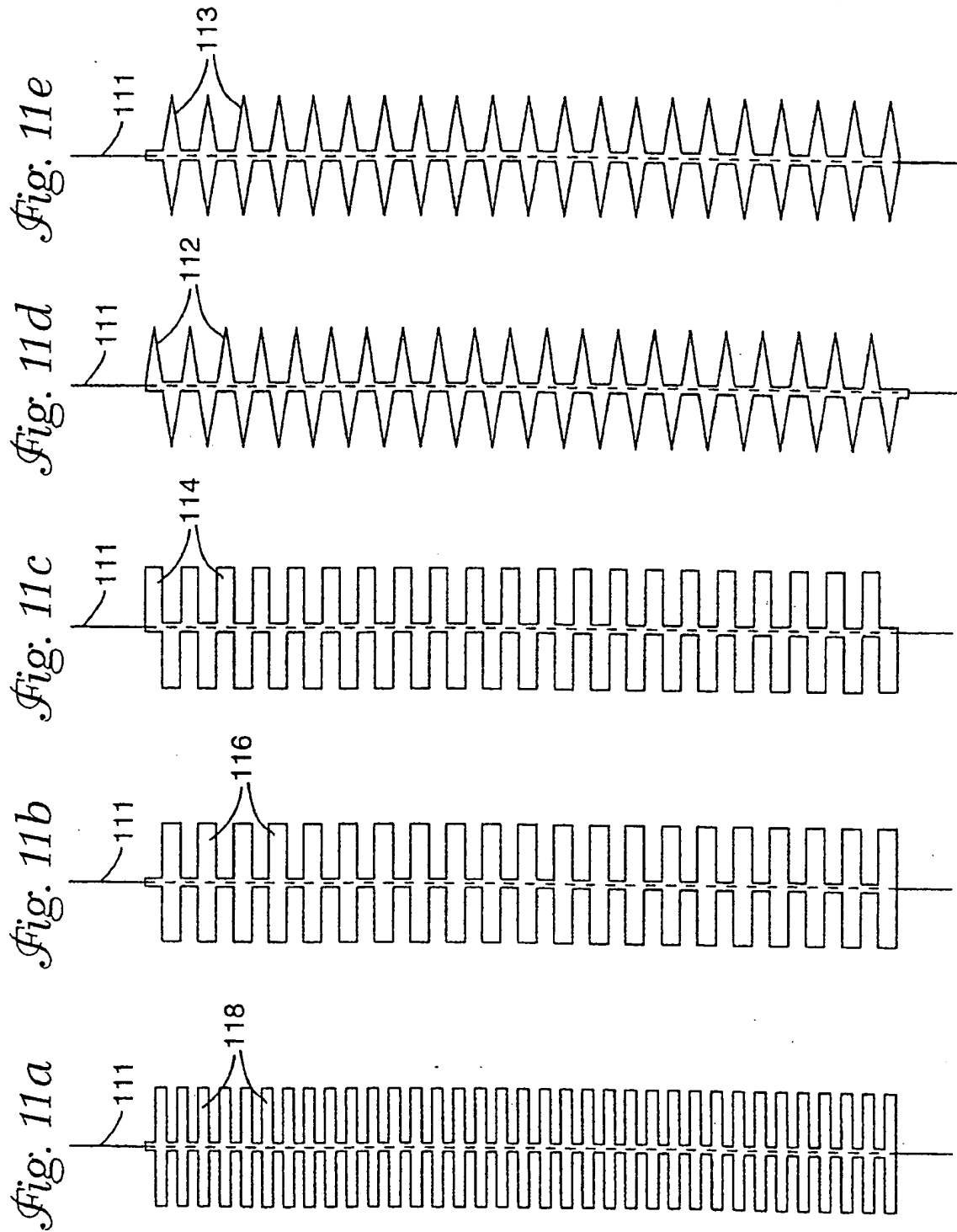
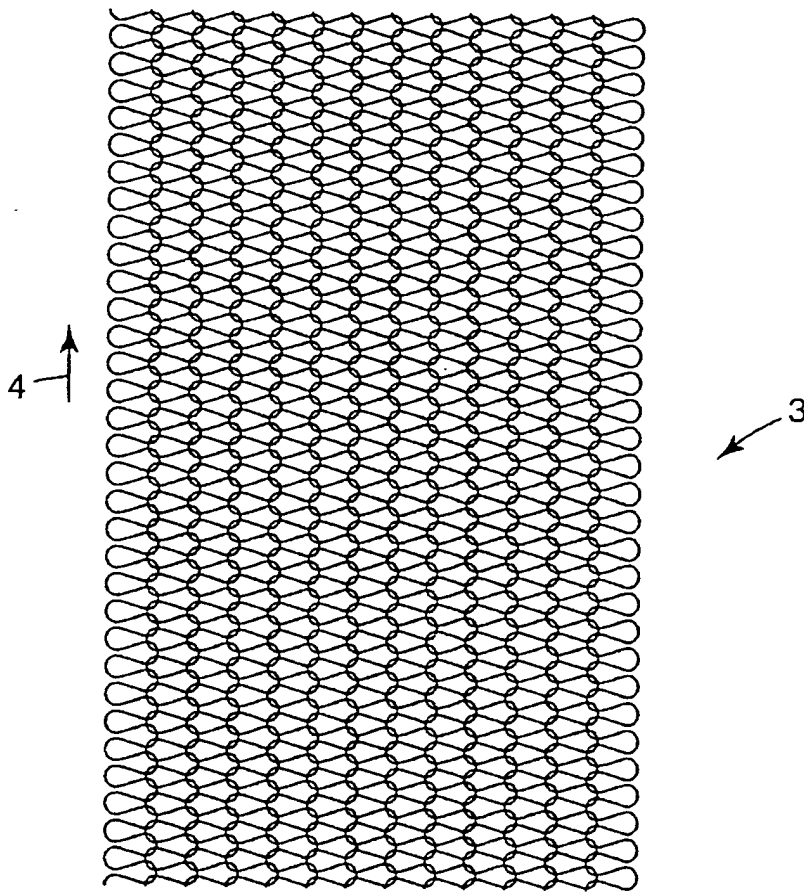
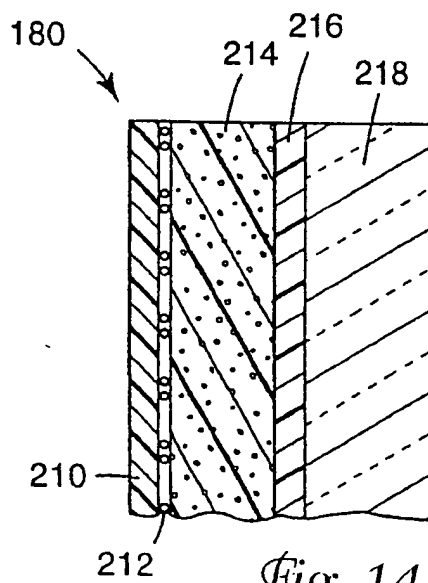
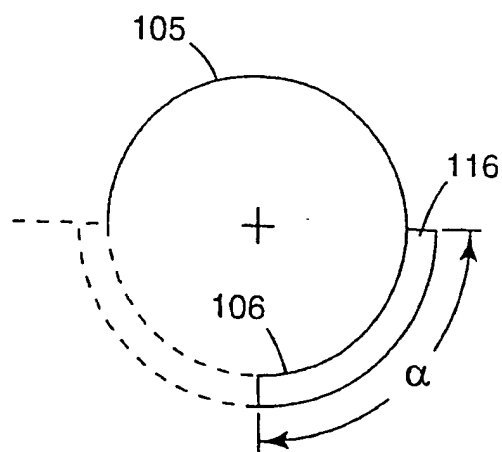
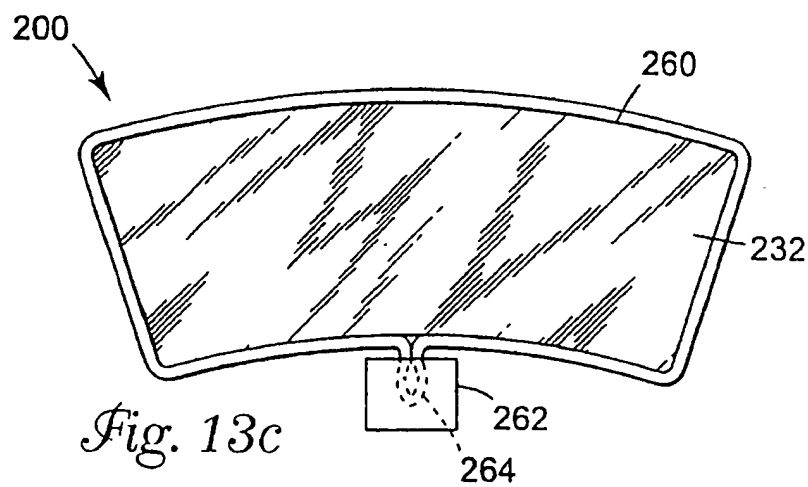
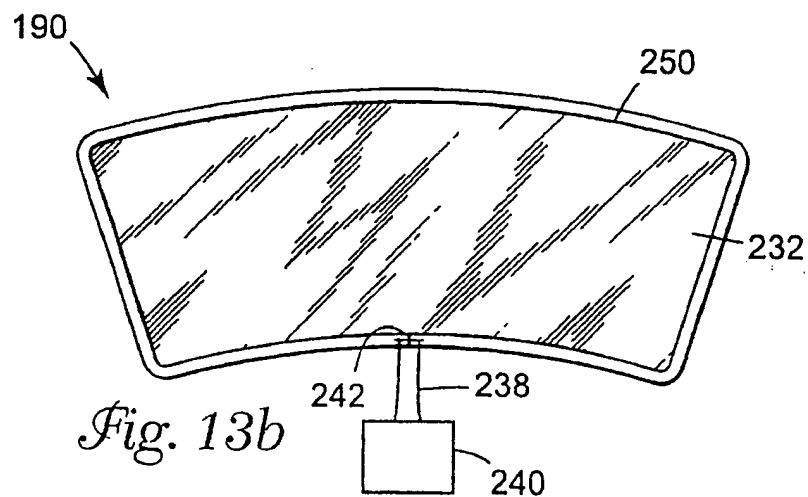
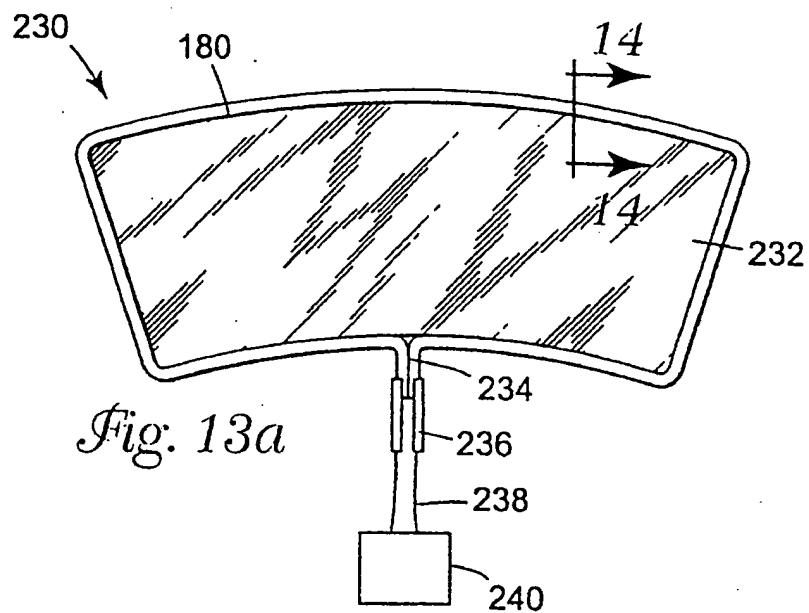


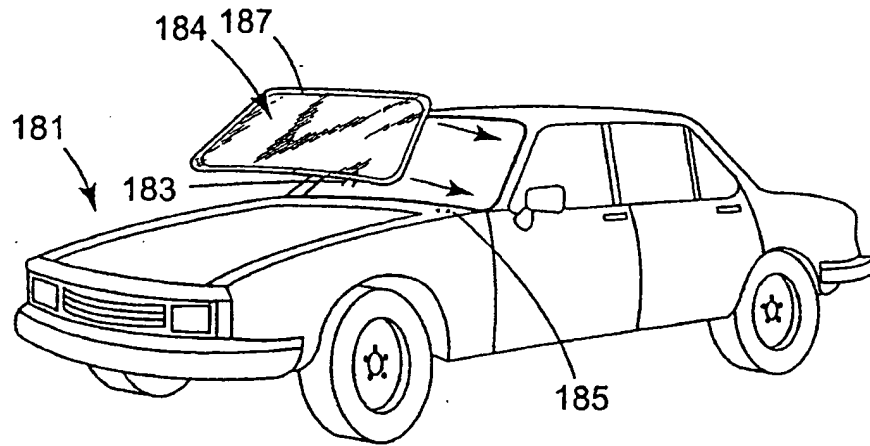
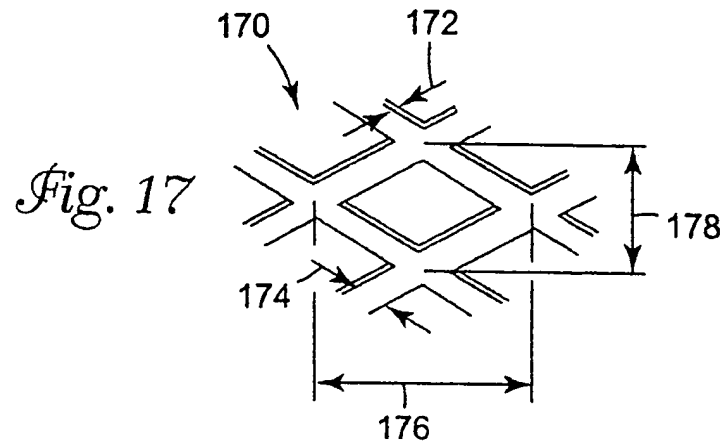
Fig. 11f

*Fig. 9a*



*Fig. 12b**Fig. 14**Fig. 15*



*Fig. 16**Fig. 17*

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 99/24255

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C09J7/02 C09J5/06 B60J10/02 B29C65/34 B32B27/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C09J B60J B29C B32B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 93 19926 A (UNITED TECHNOLOGIES CORP) 14 October 1993 (1993-10-14) cited in the application page 4, line 23 -page 5, line 28 page 7, line 24 - line 28 page 11, line 18 - line 21 figures 1,3,4	1-11
X	EP 0 128 837 A (BOUSSOIS SA ;ADHESIFS INSONORIS MODERNES (FR); LAQUES INDOCHINOISE) 19 December 1984 (1984-12-19) page 4, line 20 -page 5, line 8 figures 1-3 page 6, line 4 - line 26	1-11

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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

26 January 2000

Date of mailing of the international search report

04/02/2000

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>WO 98 18877 A (WILLE JUERGEN ALBIN ;BEIERSDORF AG (DE)) 7 May 1998 (1998-05-07) page 2, last paragraph -page 3, paragraph 1 page 3, paragraph 4 page 4, last paragraph -page 5, paragraph 3 figure 1</p>	<p>1,2,4-7, 10,11</p>
X	<p>DE 38 31 787 A (PASCAL ROGER) 6 April 1989 (1989-04-06) abstract column 3, line 43 - line 51 figures 1,5,6</p>	<p>1,2,4-7, 10,11</p>

INTERNATIONAL SEARCH REPORT

Information on patent family members

Int. Application No

PCT/US 99/24255

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